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IDA GAMMA-RAY LASER 1993 ANNUAL REPORT

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Prepared for
Ballistic Missile Defense Organization

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PREFACE

This document represents work done by the Institute for Defense Analyses in FY 1993 under Task Order T-R2-597.3, Technical Support for Innovative Concepts Program, for the Ballistic Missile Defense Organization.

ABSTRACT

This document consists of a collection of papers published in the open literature in FY 1993.

The first paper was published in *Phys. Rev. B* and describes the limitations of the Bonifacio-Lugiato model of superfluorescence.

The second paper deals with the feasibility of nuclear superfluorescence. It was presented at the Lasers '92 Conference in Houston and published in the *Proceedings*.

Paper Number 3 describes a nuclear superfluorescence model. It was presented at the Physics in Ukraine Conference in Kiev in June 1993 and published in the *Proceedings*.

Paper Number 4 is a comment on a paper published in *Phys. Rev. B*, which we found to be wrong and which stated a misleading conclusion regarding the effect of homogeneous broadening on an inhomogeneously broadened system. Our comment is followed by the author's reply, which was also published in the Comments section of *Phys. Rev. B*. We include our response to the author's reply.

CONTENTS

PAPER No. 1

CRITIQUE OF THE BONIFACIO-LUGIATO SUPERFLUORESCENCE MODEL

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Critique of the Bonifacio-Lugiato superfluorescence model

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This paper shows how the Bonifacio-Lugiato mean-field model of atomic superfluorescence, which is particularly attractive because it deals with a crystalline array of emitters, can be extended to the nuclear case. However, limits exist on the validity of certain approximations used in the development of the model. In particular, a numerical instability that increases without bound is shown to exist in the non-Markovian case, placing in doubt the application of the semiclassical theory derived for the non-Markovian case.

I. INTRODUCTION

Recent activity in the field of γ-ray lasers has focused on the possibility of observing nuclear superfluorescence (SF). The most general treatment of SF based on the Maxwell-Bloch (MB) equations[1] permits consideration of several important phenomena[2] which, while interesting only as second-order effects in atomic SF, are crucial to the understanding of nuclear superfluorescence. Consideration of emitters in a crystal lattice, which leads to special effects such as Borrmann anomalous transmission,[3] are not treated in the MB formalism. On the other hand the model that Bonifacio and Lugiato presented in 1975 (Refs. 4 and 5) deals with emitters in a crystal lattice and may be adaptable to a similar structure of nuclear emitters that radiate electromagnetic energy at wavelengths smaller than the interatomic spacing. This possibility and the quantum-mechanical basis of the BL model motivated us to revisit the early work. This paper presents some of our findings regarding the BL model.

The BL theoretical model for the behavior of superfluorescence by a system of identical two-level atoms remains an important contribution to a fundamental understanding of that phenomenon, despite the fact that the theory does not properly account for propagation or the initial quantum fluctuations of the electric-field vacuum state. For example, Polder, Schurmans, and Vrehen[6] developed a model of a similar nature without these defects; however, its derivation requires an assumption that the atoms occupy random positions, thereby ruling out its application to a crystal lattice. But not only does the BL model specifically assume that the radiating structure is a crystal, it also takes into account inhomogeneous broadening due to motion of the atoms about their periodically spaced equilibrium positions, whereas Ref. 6 neglects inhomogeneous broadening.

The crystalline structure is an important consideration when nuclear transitions resulting in photon wavelengths smaller than the interatomic spacing are responsible for the superfluorescent emission.[7] Although in deriving the BL model, the authors assume only atomic transitions for which the photon wavelengths are larger than the interatomic spacing, the associated analysis is easily adapted to the nuclear case. Section II will show that when the model is applied to the case of radiation wavelengths smaller than the interatomic spacing it predicts that superfluorescence is then only possible at wavelengths satisfying certain conditions. It turns out that these conditions are also sufficient for coupling to a Borrmann propagation mode, which has been cited as necessary for γ-ray lasing.[8]

Although the BL model approaches a classical limit in the initial stage, during which certain characteristics of the emitted pulse, such as the time delay, are determined, the treatment is quantum mechanical. Basing calculations on the more accurate quantum-mechanical equations, Sec. III, in fact, presents numerical results demonstrating that the classical approximation may not be valid until well after the radiated pulse maximum has emerged. However, Sec. IV shows that in certain circumstances a numerical instability invalidating the quantum-mechanical calculations will occur.

II. SHORT-WAVELENGTH SUPERFLUORESCENCE

If wavelengths associated with nuclear transitions are smaller than interatomic spacings, efficient radiation can only occur when conditions for Borrmann mode propagation are satisfied. Since superfluorescence is also deemed necessary for lasing in the γ-ray regime, the effect of wavelengths shorter than interatomic spacings on superfluorescence is of interest. In particular, it is of some interest to determine the extent to which conditions for superfluorescence at short wavelengths may be in conflict with those for Borrmann mode propagation.

In nuclear superfluorescence, wavelengths associated with transitions of interest will undoubtedly be smaller than the spacing of the contributing nuclei in a crystal lattice. The behavior of a similarly spaced classical periodic array of coherent sources suggests that for a potentially superfluorescent crystal, regarded as an array of identical nuclei, a first-order effect of such a short wavelength would be to limit collective radiation to certain directions. In fact, the relatively simple BL model of atomic superfluorescence has sufficient scope to confirm the existence of such an effect.

The BL superfluorescence model for two-state atoms in a rectangular crystal lattice considers only the case in

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which the emitted radiation wavelength is larger than the atomic spacing. However, this limitation is not inherent in the model, despite its simplistic treatment of propagation and other possible geometrical effects.

Reference 9 treats the case of nuclear transitions, for which the wavelength is smaller than the atomic spacing, but the paper's discussion of superfluorescence is less informative than that of BL. This is partly because the BL model uses a representation for the collective excitation operator that relates it to the lattice structure, whereas Ref. 9 uses a representation that relates the operator to the phases of the emitted photons at the lattice sites, thereby missing the effect of the regular lattice structure on the subsequent radiation.

Reference 10 gives an analysis of superradiance that, like the BL model, relates the collective excitation to the lattice structure and should therefore be capable of yielding the short-wavelength superfluorescence conditions. Because its treatment includes more geometrical detail than the BL treatment for the long-wavelength case Ref. 10 is, in fact, able to relate the spatial pattern of the radiation emitted by the collective atomic array to the characteristic shape of the active volume. The BL analysis however, is restricted to the case in which the active volume is a thin needle.

On the other hand, Ref. 10 shows that the needle shape provides the maximum gain, albeit in a single direction. Thus, to the extent that it is possible to implement, the needle is presumably the preferred shape for a superradiant structure. Therefore, it seems useful to consider the consequences of the BL model for the short-wavelength radiation that characterizes the nuclear case.

The interaction Hamiltonian originally introduced by BL has the form

$$H_I(t)=\frac{i\hbar}{\sqrt{V}}\sum_{j=1}^{N}\sum_{\mathbf{k}} g_{\mathbf{k}}\left\{a_{\mathbf{k}}^{\dagger} r_j^{-}\exp[i(\omega_0-\omega_j)t-i\mathbf{k}\cdot\mathbf{x}_j]-\text{H.c.}\right\}, \tag{1}$$

where $\mathbf{k}$ is the emitted photon wave vector for a particular mode, $\mathbf{x}_j$ is the position vector of the jth lattice site, $a_{\mathbf{k}}^{\dagger}$ is the corresponding photon creation operator, ω_0 is a mean reference frequency, and r_j^{-} is the deexcitation operator for the atom at the jth lattice site. The corresponding excitation operation is r_j^{+}. BL also define the reciprocal-lattice vectors α, each component of which has the form $(2\pi/L)n$, $n=0,1,\ldots,N-1$, in which L is the corresponding lattice dimension. They define the collective excitation and deexcitation operators $R^{\pm}(\alpha)$ as functions of α by

$$R^{\pm}(\alpha)=\sum_{j=1}^{N} r_j^{\pm}\exp(\pm i\alpha\cdot\mathbf{x}_j), \tag{2}$$

from which it can be shown that

$$r_j^{\pm}=\frac{1}{N}\sum_{\alpha} R^{\pm}(\alpha)\exp(\mp i\alpha\cdot\mathbf{x}_j). \tag{3}$$

On substituting (3) into (1) it follows that

$$H_I(t)=\frac{i\hbar}{\sqrt{V}}\sum_{\alpha}\sum_{\mathbf{k}} g_{\mathbf{k}}[a_{\mathbf{k}}^{\dagger}R^{-}(\alpha)f^{*}(\mathbf{k}-\alpha,t)-\text{H.c.}], \tag{4}$$

where

$$f(\eta,t)=\frac{1}{N}\sum_{j=1}^{N}\exp(i\eta\cdot\mathbf{x}_j)\exp[i(\omega_j-\omega_0)t]. \tag{5}$$

The time-dependent factors in (5) are separated out on the assumption that they can be replaced by an average over all atoms and that, because the frequencies ω_j associated with the individual atoms are uncorrelated, they can be removed from $f(\eta,t)$ and subsumed in the coupling constant $g_{\mathbf{k}}$, which then becomes time dependent. This time dependence of $g_{\mathbf{k}}$ is a way of introducing inhomogeneous broadening.

At this point, BL introduce the assumption that the atomic spacing, given by

$$d=L/N,$$

is much smaller than the photon wavelength, which implies that the function $f(\eta)$ (wherein the time dependence is no longer indicated for the reason just noted) is large when η vanishes and is otherwise small. However, if this assumption is not made then $f(\eta)$ will be large whenever $\eta\cdot\mathbf{x}_j=2\pi\nu_j$, $\nu_j=0,\pm1,\ldots$ and otherwise small.

The interaction that results in photon emission when cooperative deexcitation of the atoms takes place can only occur when $f(\mathbf{k}-\alpha)$ in (4) is not negligible for some value of $\mathbf{k}$ and α, i.e., when

$$(\mathbf{k}-\alpha)\cdot\mathbf{x}_j=2\pi\nu_j, \tag{6a}$$

where ν_j is in the set: $0\pm1,\ldots$, which implies that

$$d/\lambda-n/N=\nu,\quad n=0,1,\ldots,N-1, \tag{6b}$$

where ν is in the set: $0,\pm1,\ldots$. In (6) it is tacitly assumed that the propagation direction is along a narrow, needle-shaped active volume, in which only a single-plane-wave photon mode is supported. More generally, λ should be interpreted as λ_j, the photon wavelength divided by the component of the unit vector in the propagation direction along the lattice direction defined by the $\mathbf{x}_j$.

It follows from 6(b) that the possible values of ν must be non-negative. Thus,

$$\lambda_j=L/(N\nu_j+n_j),\quad 0<n_j<N,\ \nu_j\geq 0. \tag{7}$$

On setting $\nu_j=0,1,\ldots$ in succession it follows from (7) that for $\nu_j=0,\lambda_j>d$, for $\nu_j=1,d\geq\lambda_j\geq d/2$, for $\nu_j=2,d/2\geq\lambda_j>d/3$, etc. That is, for any given photon wavelength, one and only one value of ν exists that will satisfy the required condition.

Reference 9 gives as the condition for a Borrmann mode the relation

$$|\mathbf{k}-\alpha|=|\mathbf{k}| \tag{8}$$

for some reciprocal-lattice vector α. To satisfy (8) it is

sufficient that $k=\alpha'$ for some reciprocal-lattice vector[11] α', a condition that (7) implies. That is, the condition implied by the BL model for superradiance in a crystal is sufficient for coupling to a Borrmann mode, the propagation direction of which will be determined by the ratio of the wavelength λ to the atomic spacing d in accordance with (7).

III. VALIDITY OF THE SEMICLASSICAL APPROXIMATION

BL show that their model implies two basic conservation laws. One preserves the balance between emitted radiation and stored energy, and the other preserves the Dicke cooperation eigenvalue defined in terms of the time-varying atomic dipole polarization and population inversion states.

The first law has the form

$$\frac{d}{dt}\left\{\sum_{\alpha=k_0-k_0}[\langle A^\dagger(\alpha)A(\alpha)\rangle(t)]+\langle R_3\rangle(t)\right\} = -2K\sum_{\alpha=k_0-k_0}[\langle A^\dagger(\alpha)A(\alpha)\rangle(t)]\,, \quad (9)$$

where $A(k_0)$ is the resonant mode of the internal field, k_0 is the vector wave number at resonance, R_3 is the population inversion, and K (given by $c/2L$, where L is the axial length of the active volume and c is the velocity of light) is the reciprocal maximum round trip transit time of photons in the active volume. The brackets $\langle\ \rangle$ refer, as usual, to the expectation value of the operator that they enclose, and parentheses ( ) to a functional dependence on the independent variable that they enclose. The second law, in which R^+ and R^- are collective dipole moment operators, has the form

$$\frac{d}{dt}\left\{\sum_{\alpha=k_0-k_0}[\langle R^+(\alpha)R^-(\alpha)\rangle(t)] + \langle R_3^2\rangle(t)-\langle R_3\rangle(t)\right\}=0\,. \quad (10)$$

Any Hamiltonian system would imply the first law. The second is analogous to and formally identical with the standard conservation of angular momentum (resulting in this case from a collection of pure spin states) when the total angular momentum is identified with the Dicke cooperation eigenvalue associated with a collective total angular momentum (spin) operator R, and the angular momentum vector components are identified with R_3 and the real and imaginary parts of R^+.

The conservation laws are therefore physically reasonable in their own right. In fact, they appear to be quite general and could be regarded as essential requirements for any model based on the collective behavior of identical two-state atoms.

From their general master equation, specialized to the case of two identical, independent, single-resonant modes, BL also derive another equation involving the expectation values of the atomic and electromagnetic field operators:

$$\langle\ddot{R}_3\rangle(t)+\left[K+\frac{1}{2T_2^*}\right]\langle\dot{R}_3\rangle(t)=-\frac{2g_0^2}{v}e^{-t/T_2^*}\left\{\sum_{\alpha=k_0-k_0}[\langle R^+(\alpha)R^-(\alpha)\rangle(t)+2\langle A^\dagger(\alpha)A(\alpha)R_3\rangle(t)]\right\}, \quad (11)$$

where g_0 is the coupling constant at resonance, v is the volume of the active region, and T_2^* is a time constant due primarily to inhomogeneous line broadening. The unknown quantities appearing in Eq. (11) and the two conservation laws (9) and (10) are the photon number expectation $\langle A^\dagger A\rangle$, the atomic inversion expectation $\langle R_3\rangle$, the photon number/atomic inversion correlation $\langle A^\dagger AR_3\rangle$, and the fluctuations $\langle R^+R^-\rangle, \langle R_3^2\rangle$ of the atomic dipole/inversion vector components.

Equations (9), (10), and (11) derived from the BL model do not form a complete set of relations for all of the explicitly involved quantities that must be taken as independent in a quantum-mechanical treatment. However, in the classical approximation the relations

$$\langle R_3^2\rangle=\langle R_3\rangle^2\,,$$

$$\langle A^\dagger AR_3\rangle=\langle A^\dagger A\rangle\langle R_3\rangle\,,$$

hold, and they reduce the number of unknowns to 3. Then (9), (10), and (11) reduce to a differential equation, similar to that derived from classical mechanics for the motion of a pendulum, and a corresponding energy relation which, together, do form a closed system:

$$\sum_{\alpha=k_0-k_0}\langle A^\dagger(\alpha)A(\alpha)\rangle(t)=\frac{v}{4g_0^2}[\dot\phi(\tau)]^2e^{t/T_2^*}\,,$$
$$\ddot\phi(t)+\left[K+\frac{1}{T_2^*}\right]\dot\phi(t)-\frac{g_0^2N}{v}e^{-t/T_2^*}\sin\phi(t)=0\,, \quad (12)$$

where $\phi(t)$ is a modified Bloch angle defined by

$$\langle R_3\rangle(t)=[1+N\cos\phi(t)]/2\,. \quad (13)$$

With relations (12) and (13), to calculate the population inversion and the emitted electromagnetic radiation, given by

$$I(t)=\frac{Kv}{2g_0^2}[\dot\phi(t)]^2e^{t/T_2^*} \quad (14)$$

as functions of time, is comparatively simple.

Thus, where the classical decorrelations are valid, the BL model gives a firm footing to a numerically tractable differential equation, permitting straightforward calculations of the most important quantities associated with

superfluorescence. The equation includes line broadening and accounts fully for the effects of atomic stimulation by the local field, which is due to spontaneous emission by the initially excited atoms.

Further on, this section will present some numerical results obtained from the BL model with the aid of approximations from[4,5] that, unlike the classical, preserve first-order quantum-mechanical effects. Those results indicate the presence of large quantum fluctuations during the time period when most of the radiant pulse energy is emitted. This is somewhat disturbing because the validity of the classical approximation over any time interval should depend on quantum fluctuations being small enough to be neglected during the interval.

In Ref. 12 Bonifacio, Schewendiman, and Haake report a similar finding derived from an earlier, more primitive version of the BL model:[13] one that does not include stimulation effects. Physically, the earlier model (which is Markovian and is a limiting form of the more sophisticated version as the ratio of the cooperation time to the photon propagation time becomes large) differs from that of Ref. 5 by virtue of the fact that the radiated photons leave the active volume before they can interact with the atoms. As a result, they follow the atomic state changes adiabatically and do not produce a ringing effect in the emitted pulse.

With the earlier model, the authors are able to show by direct calculation[12] that quantum fluctuations are large when the atomic system is totally inverted initially. But when the active population is sufficiently large and initially less than totally inverted, their calculations show that the fluctuations are small. They also demonstrate that, consistent with this result, the classical approach is valid whenever the initial atomic system is less than totally inverted.

Although the results in Ref. 12 appear to validate the use of the classical approximation to predict spontaneous cooperative radiation whenever the atoms are not totally inverted initially, the more sophisticated Ref. 5 model does not necessarily lead to the same conclusion when non-Markovian effects are important. This puts the validity of the semiclassical approach in doubt during the time period when most of the radiation takes place.

Unfortunately, the region of validity of the Ref. 5 approximations, which take into account quantum-mechanical effects, is itself uncertain, at least for the case in which the process is non-Markovian. Thus, calculations based on those approximations cannot be used directly to assess the accuracy of the semiclassical approach over the questionable time period.

To take into account quantum fluctuations in the non-Markovian case, BL make two approximations. One is the Born approximation which, without some additional step such invoking the semiclassical decorrelations, does not lead directly to a closed system of equations for expectation values of photon and atomic operators.

BL introduce the Dicke states $|n,m\rangle$, which are eigenvectors of the photon number operator $A^{\dagger}A$ and the inversion operator R_3, so that

$$A^{\dagger}A|n,m\rangle = n|n,m\rangle ,$$

$$R_3|n,m\rangle = m|n,m\rangle .$$

In terms of an operator $\Phi(t)$, which is a projection of the density operator of the system that leaves the atomic observables unaltered, BL define the occupation probabilities

$$p(m,t)=\sum_n \langle n,m|\Phi(t)|n,m\rangle ,$$

the photon number expectations

$$N(m,t)=\sum_n \langle n,m|\Phi(t)|n,m\rangle ,$$

and the quantities

$$L(m,t)=\mathrm{Re}\sum_n [(n+1)(n+2)]^{1/2} \times \langle n+2,m-2|\Phi(t)|n,m\rangle .$$

In the Dicke state representation, the Born approximation leads at first to a finite system of integrodifferential equations for the occupation probabilities $p(m,t)$ of the Dicke state basis vectors $|r,m\rangle$:

$$\dot{p}(m,t)=-\frac{2g_0^2}{v}\int_0^t ds\, e^{-(t+s)/2T_2^*-K(t-s)} \times\{g(m)p(m,s)-g(m+1)p(m+1,s)+[g(m)+g(m+1)]N(m,s) -g(m+1)N(m+1,s)-g(m)N(m-1,s)+g^{1/2}(m)g^{1/2}(m-1)L(m,s) -2g^{1/2}(m)g^{1/2}(m+1)L(m+1,s)+g^{1/2}(m+1)g^{1/2}(m+2)L(m+2,s)\} , \quad (15)$$

where

$$g(m)=\begin{cases}(\frac{1}{2}N'+m)(\frac{1}{2}N'-m+1) & \text{for } -\frac{1}{2}N'\le m\le \frac{1}{2}N' ,\\ 0, & \text{otherwise} ,\end{cases}$$

and N' is the number of emitters. Unfortunately, the number of unknown quantities to be determined from Eq. (15) is larger than the number of equations in the system. When the authors enlarge the system to include time derivatives of the unknowns other than the Dicke state occupation probabilities, the resulting set of equations is still not closed because it contains new unknowns, consisting of higher-order moments of the photon and atomic operators. Obviously,

repeating the process indefinitely will result in an infinite hierarchy of (finite) systems of equations, involving moments of ever increasing order.

BL observe that a simple way of getting a closed system is to drop the photon number expectation values (but retain their derivatives on the left-hand side) and all second-order moments from the first set of equations in this hierarchy that involve time derivatives of just the Dicke state occupation probabilities and the photon number expectations. However, that procedure should be valid only under conditions that would justify substituting the earlier Markovian model. It would therefore add nothing new, serving only to verify that their earlier model is a limiting case of the more general non-Markovian model.

Their next step, then, is to keep the equations for which the left-hand side involves time derivatives of the Dicke state occupation probabilities and the photon expectation values and retain the Dicke state occupation probabilities and the photon expectation values on the right-hand side, but to drop all second-order and higher moments. The result is a larger, but still closed, system:

$$\dot{p}(m,t)=-\frac{2g_0^2}{v}\int_0^t ds\, e^{-K(t-s)-(t+s)/2T_2^*}$$

$$\times\{g(m)p(m,s)-g(m+1)p(m+1,s)+[g(m)+g(m+1)]N(m,s)$$

$$-g(m+1)N(m+1,s)-g(m)N(m-1,s)\}\ , \tag{16a}$$

$$\dot{N}(m,t)=-2KN(m,t)+\frac{2g_0^2}{v}\int_0^t ds\, e^{-K(t-s)-(t+s)/2T_2^*}$$

$$\times\{g(m+1)[p(m+1,s)+N(m+1,s)-N(m,s)]\}\ . \tag{16b}$$

Significantly, the equations resulting from this approximation imply a relationship between atomic and photon operator expectation values that is also implied by the exact (i.e., with no approximations) operator equations of the model. Therefore, the three previously discussed equations, (9), (10), and (11), follow from (16). In addition, (16) guarantees conservation of probability: The sum of the Dicke state occupation probabilities satisfying (16) must remain constant over time. This result follows from the identity

$$\sum_{m=-\infty}^{\infty} g(m+1)F(m+1)=\sum_{m=-\infty}^{\infty} g(m)F(m)\ ,$$

in which $F(m)$ can be any function.

Quantum-mechanical fluctuations are responsible for the triggering of the superfluorescent pulse from the inverted population. It has been argued that when these fluctuations are small, classical solutions give good approximations to the superfluorescent pulse emission and that these fluctuations are small at the maximum emission rate.[13–15]

To check this assumption and study the characteristics of solutions obtained from Eq. (16) we calculated the emitter pulse given by

$$I(t)=\sum_m N(m,t) \tag{17}$$

and the variance defined as

$$\langle\langle R_3\rangle\rangle=\langle R_3^2\rangle-\langle R_3\rangle^2$$

$$=\sum_m m^2 p(m,t)-\left\langle\sum_m mp(m,t)\right\rangle^2\ . \tag{18}$$

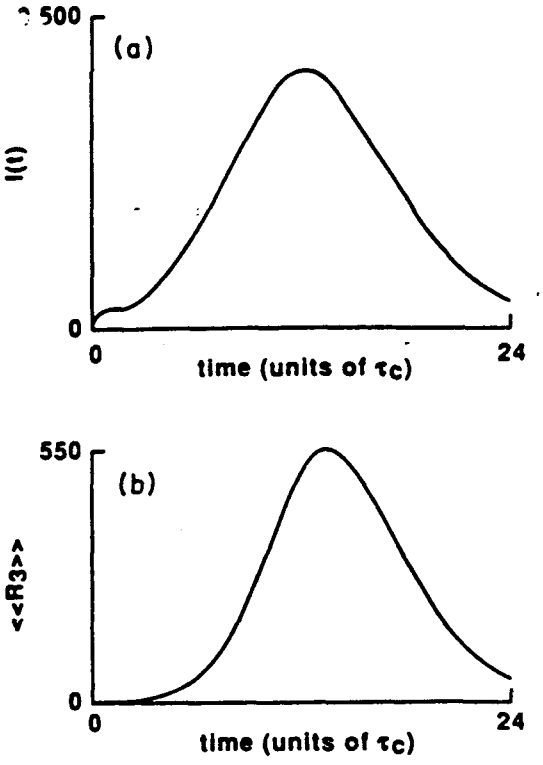

FIG. 1. SF pulse intensity (a) and variance (b) calculated for the system with $N=100$ ($-50\le m\le 50$) and $K=10$. The initial condition is the totally inverted state with the probability $P(m,0)=0$ for all m except for $m=50$ for which $P(50,0)=1$.

For the calculations we will discuss here we assumed 100 particles so that $N=100$ and m varies from -50 to $+50$. We assumed different initial conditions for the probabilities $P(m,t)$ and values of K to obtain both pure and oscillatory SF.[16] In our calculations we measured time in units of the cooperation time $\tau_c=(1/g_0KV/N)^{1/2}$. Figure 1 shows the result of our calculations under pure SF conditions with $K=10$. The initial probabilities are $P(m,t)=0$ for all m except $m=50$, which give a fully inverted state. Note that the variance is maximum when the pulse is being emitted [compare Figs. 1(a) and 1(b)]. These results indicate that the quantum fluctuations are not necessarily negligible during the course of the emission process. Also note in Fig. 2 the shift of the state population from the inverted state [right side in (a)] at early times to the broader distribution at intermediate times (d), (e), and (f) and finally the ground-state population at higher times (g) and (h).

When we attempt to run the same calculation with $K<2\sqrt{2}$ an instability develops: The intensity oscillates wildly until the calculation blows up. The violent oscilla-

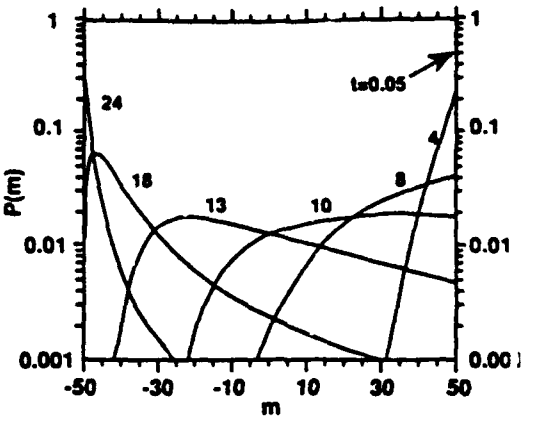

FIG. 2. Probability distribution, $P(m,t)$ at different times for the states $-50\le m\le 50$, used in the calculation of the SF intensity and variance shown in Fig. 1. The times at which the probability $P(m,t)$ is shown are $t=0.05$, 4, 8, 10, 13, 18, and 24 in (a)–(h), respectively.

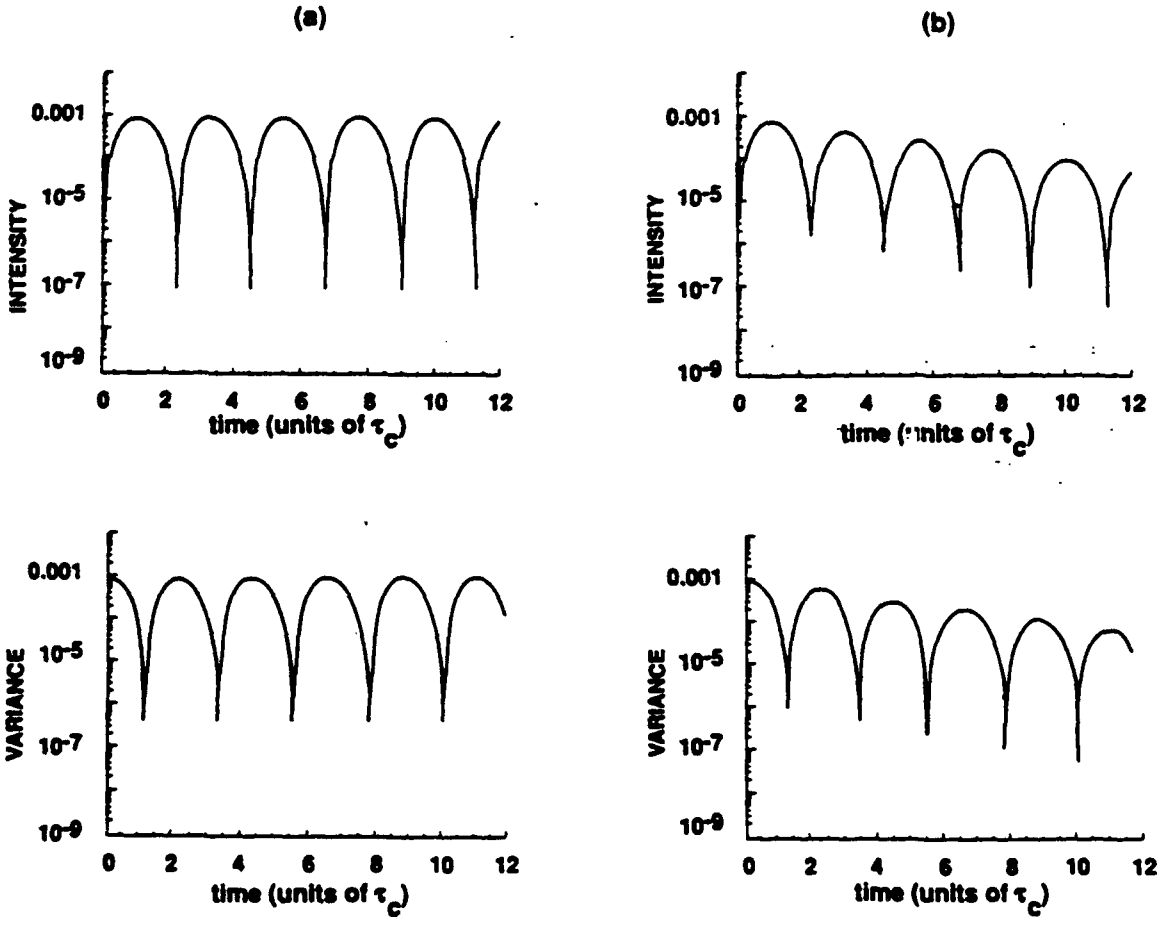

FIG. 3. Radiative pulse and quantum fluctuations (variance) for different K values which are 0.0269 units in (a), 0.269 in (b), 0.8 in (c), and 2.69 in (d). The initial distribution for (m) is Poisson with $P_c=10^{-5}$ peaked at $m=-50$ and $N(m)=0$.

tions and blow up can be reduced by starting with the population closer to the ground state, as shown in Fig. 3, where the Poisson distribution is peaked at the ground state $M=-50$. In Fig. 3 for K values from 0.0269 to 2.69, below the critical value of $2\sqrt{2}$ the emitted pulses remain stable and the variance is positive. This is not the case shown in Fig. 4 when the initial distribution is peaked at $m=-46$ [$P(m,t)=1$ for $m=-46$ and zero otherwise] and $K=0.0269$, the intensity goes slightly negative and the variance oscillates between positive and negative values. With the same initial distribution but with higher $K=2.69$ (still less than $2\sqrt{2}$) the single pulse appears and the variance stays positive as shown in Fig. 5. In Sec. IV we examine the source of the instabilities and the conditions for their occurrence. We also discuss a closed-form analytical solution obtained for a system with two particles.

IV. NUMERICAL INSTABILITY IN THE BL APPROXIMATION

Because the approximate equations (16) yield the exact basic conservation laws of the BL model, it might be expected that their solutions would be physically well behaved. However, numerical calculations indicate otherwise.

After a certain time interval, before the emitted pulse reaches its maximum amplitude, quantities derived from the approximate solution become nonphysical in at least two respects. First, although conservation of total probability is still satisfied, individual Dicke state occupation probabilities become negative. Second, although conservation of energy is still satisfied, individual Dicke state photon number expectation values also become negative.

The case in which the number of atoms in the active volume is limited to two is simple enough to be treated in detail analytically if inhomogeneous line broadening is neglected. An investigation of it using the Laplace transform reveals that all states below the maximum Dicke occupation number (which is two in the case considered) exhibit a resonance phenomenon; i.e., some of the transformed solution functions have double poles. In the inverse transform domain such a function, which would otherwise be exponential or trigonometric, must have the time variable as a factor.

In the absence of double poles, when parameters are

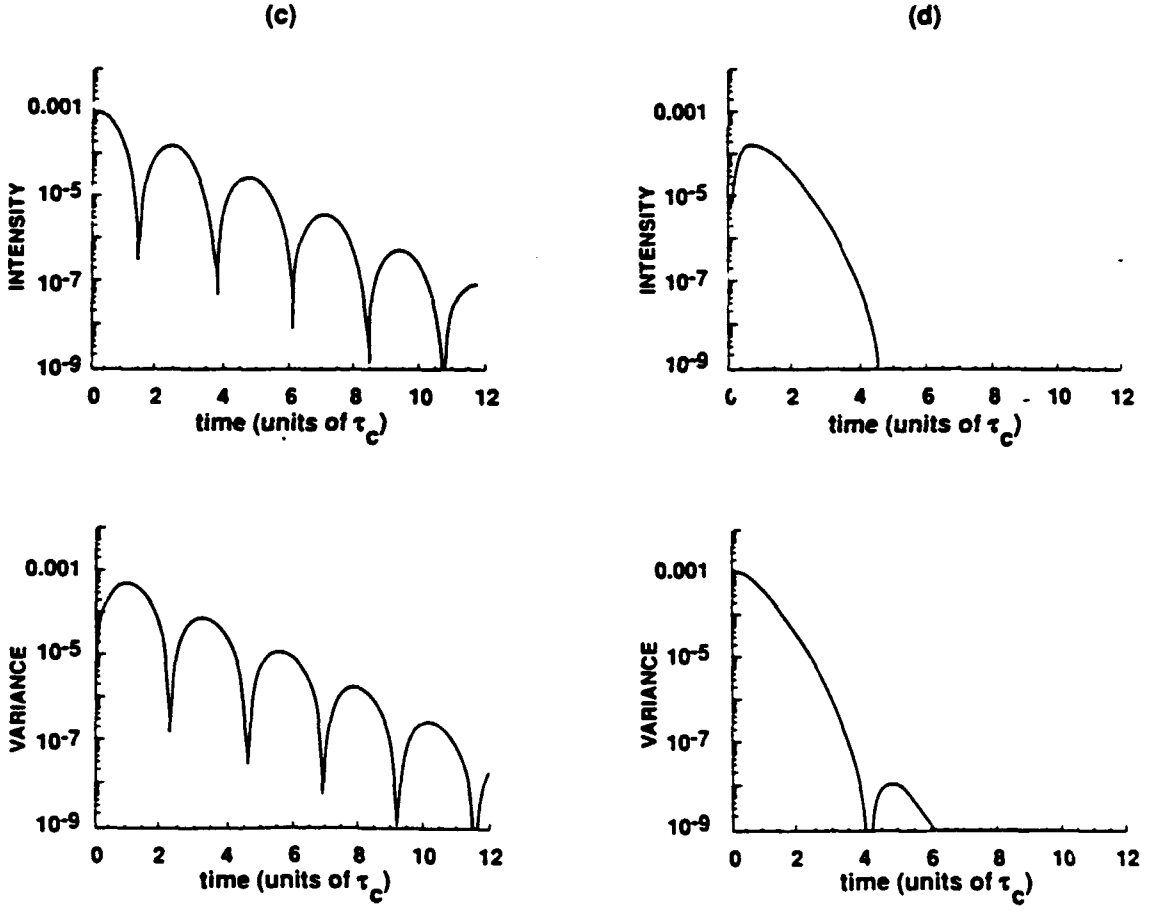

FIG. 3. (*Continued*).

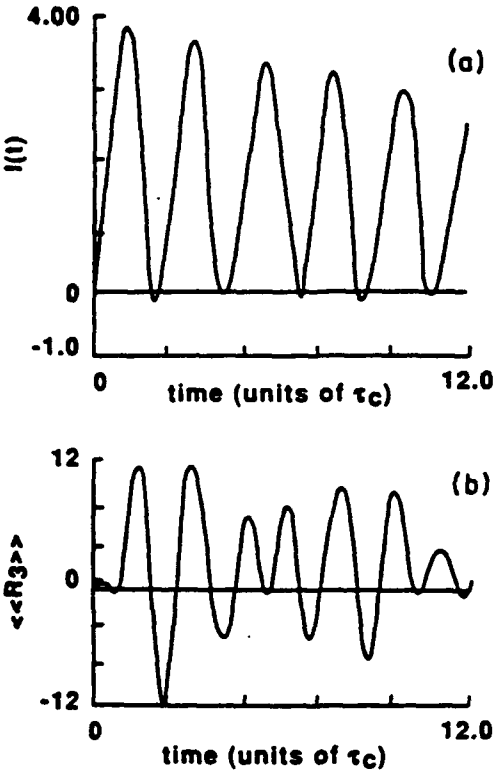

FIG. 4. Pulse (a) and variance (b) for the initial conditions $p(5,0)=1$, $p(m,0)=0$, $m\neq 5$, $N=100$, and $K=0.0269$.

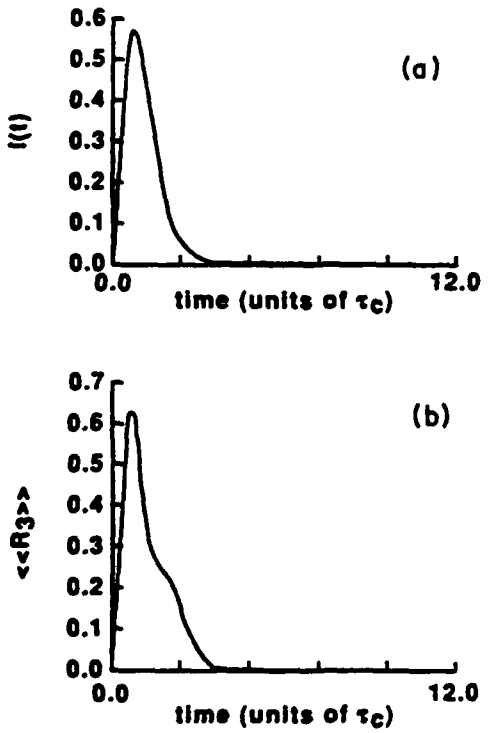

FIG. 5. Pulse (a) and variance (b) for the initial conditions $p(5,0)=1$, $p(m,0)=0$, $m\neq 5$, $N=100$, and $K=2.69$.

such that ringing does not occur, the solution functions decay exponentially with time. When the converse is true, i.e., parameters are such that ringing does occur, the solution functions have factors that are linear combinations of trigonometric functions of time. At least in the second case, the additional time variable factor imposed by a double pole guarantees increasingly larger oscillations with linearly increasing amplitudes that must eventually produce negative expectation values.

In principle, a similar analysis for an arbitrary number of atoms could be carried out in the same way, using the Laplace transform, since the equations can be treated recursively in pairs, no matter what the total number of atoms may be. Because of the symmetry of the inversion operator eigenvalues about zero, the coefficients due to the lower Dicke states will always produce double poles for the transformed solution functions, just as in the case of two atoms.

In fact, in Ref. 12 the authors make the same observation in connection with the earlier Markovian model, i.e., that due to this coefficient symmetry, double poles must always occur in the transformed solution functions. However, in Ref. 12 the remark is made in passing, without noting the consequence that, in approximate solutions for a more general model, such resonances may lead to physically impossible negative expectation values.

Equations (16) are approximate relations for the occupation probabilities $p(m,t)$ of the atomic Dicke states $|r,m\rangle$ and the corresponding photon expectation values $N(m,t)$ of the radiating electromagnetic field, given a particular value $(N'/2)(N'/2+1)$ for the atomic cooperation eigenvalue. For the sake of an analytic example we now take the "inhomogeneous broadening" decay time T_2^* to be infinite. Time is measured in units of the cooperation time τ_c; thus, the quantity $2g_0^2/v$ becomes $4/N'$.

Along with Eq. (16) the standard initial conditions

$$N(m,0)=0\,,$$
$$p\left[\frac{N'}{2},0\right]=1\,, \tag{19}$$
$$p(m,0)=0,\quad m<\frac{N'}{2}$$

will be assumed here. These conditions imply that initially all N' atoms are excited and the photon field is in the vacuum state. Together with (16) they imply that $N(N'/2,t)\equiv 0$.

Also, with neglect of T_2^*, the solution of (16) can be found by means of the Laplace transform. Then, setting $\overline{p(m,z)}$ and $\overline{N(m,z)}$ for the Laplace transforms of $p(m,t)$ and $N(m,t)$, (16) is equivalent to

$$\left[z+2K+\frac{4}{z+K}\right]\overline{N\left[\frac{N'}{2}-1,z\right]}-\frac{4}{z+K}\overline{p\left[\frac{N'}{2},z\right]}=0\,,$$
$$-\frac{4}{z+K}\overline{N\left[\frac{N'}{2}-1,z\right]}+\left[z+\frac{4}{z+K}\right]\overline{p\left[\frac{N'}{2},z\right]}=1\,,$$

for the highest m values and

$$\left[z+2K+\frac{\gamma_m}{z+K}\right]\overline{N(m-1,z)}-\frac{\gamma_m}{z+K}\overline{p(m,z)}$$

$$=\frac{\gamma_m}{z+K}\overline{N(m,z)}\,, \tag{20}$$

$$-\frac{\gamma_m}{z+K}\overline{N(m-1,z)}+\left[z+\frac{\gamma_m}{z+K}\right]\overline{p(m,z)}$$

$$=-\left[\frac{\gamma_m+\gamma_{m+1}}{z+K}\right]\overline{N(m,z)}$$

$$-\frac{\gamma_m+1}{z+K}\overline{N(m+1,z)}-\frac{\gamma_m+1}{z+K}\overline{p(m+1,z)}$$

for the rest, where

$$\gamma_m=\frac{4g(m)}{N'}\,. \tag{21}$$

The quantities $N(m-1,z)$ are naturally matched with the quantities $p(m,z)$ in Eq. (20) in the recursive pairs, the quantity $N(N'/2,t)$ already having been found to be zero identically. The determinant, Det(z), of the matrix on the left side of Eq. (20) is given by

$$\mathrm{Det}(z)=\left[z+2K\frac{\gamma_m}{z+K}\right]\left[z+\frac{\gamma_m}{z+K}\right]-\frac{\gamma_m{}^2}{(z+K)^2}$$

$$=z^2+2Kz+2\gamma_m\,. \tag{22}$$

Accordingly, the inverse matrix, which can be used to solve Eq. (20), is given by

$$A^{-1}=\begin{bmatrix}\dfrac{z^2+Kz+\gamma_m}{\mathrm{Det}(z)}, & \dfrac{\gamma_m}{\mathrm{Det}(z)}\\[2ex] \dfrac{\gamma_m}{\mathrm{Det}(z)}, & \dfrac{z^2+3Kz+2K^2\gamma_m}{\mathrm{Det}(z)}\end{bmatrix}. \tag{23}$$

The zeros of Det(z) determine whether the solution pair $N(m,t)$ and $p(m,t)$ for a given value of m oscillate with a natural frequency that does not depend on the right side of Eq. (20). If the zeros are both real, those quantities do not have a natural oscillation frequency; if the zeros are complex (in which case they occur in conjugate pairs) they have a natural oscillation frequency ω given, except for sign which is conventionally positive, by the imaginary part of either zero.

The zeros are given by

$$z=-K\pm\sqrt{K^2-2\gamma_m}=-K\pm\sqrt{K^2-8g(m)/N'}\,. \tag{24}$$

The condition for a natural oscillation frequency corresponding to a given m is therefore

$$K<2\sqrt{2g(m)/N'}\,. \tag{25}$$

The definition of the $g(m)$, along with (25), together imply that ringing occurs in the system if and only if[17]

$$K<2\sqrt{2}\,.$$

This is a more precise condition for the existence of a non-Markovian stimulation effect than the Ref. 5 condition, which in terms of the present notation would be $K\sim 1$.

If oscillation at a natural frequency ω occurs for some value m' of m, then, since the corresponding $N(m'-1,t)$ and $p(m',t)$ will contribute to the sources in the equations for $N(m'-2,t)$ and $p(m'-1,t)$, each of those quantities will also have a term oscillating with the frequency ω, etc. Thus, if there is a natural frequency again equal to ω for some $m<m'$, the corresponding $N(m-1,t),p(m,t)$ will exhibit resonant vibrations.

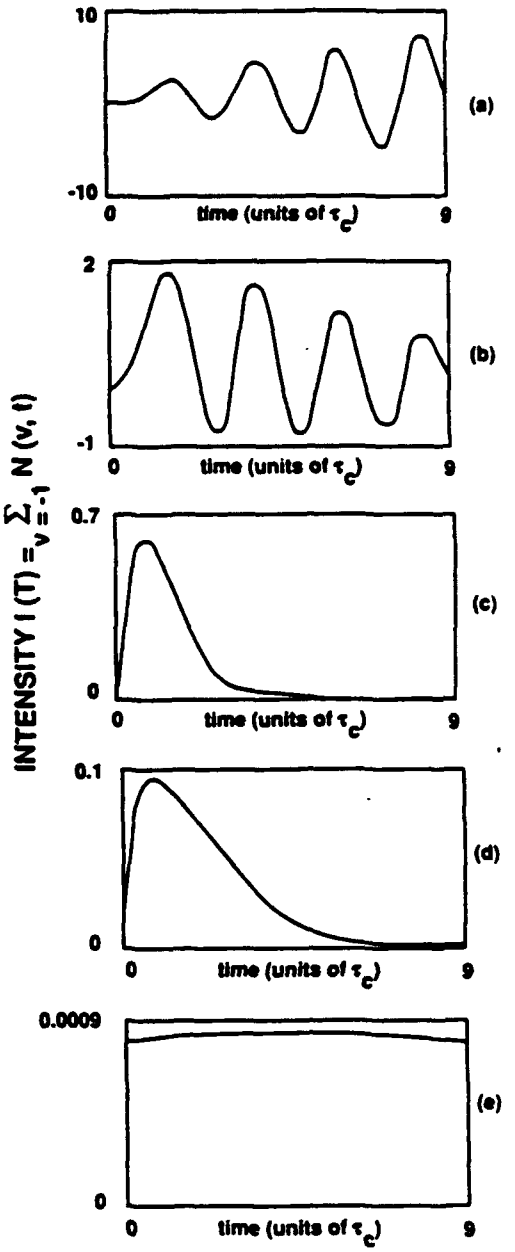

FIG. 6. SF pulses given by $I(t)=\sum_m^{N'}N(m,t)$ for $N'=2$ calculated from Eq. (32). In (a) $K=0.03$, (b) $K=0.3$, (c) $K=3$, (d) $K=5$, (e) $K=50$.

It is evident from (24) that this can occur if and only if

$$g(m')=g(m) .$$

Since

$$g\left[\frac{N'}{2}-\mu\right]=(\mu+1)(N'-\mu) ,$$

$$g\left[\frac{N'}{2}-\mu\right]=g\left[\frac{N'}{2}-\nu\right] ,$$

if and only if

$$N'=\mu+\nu+1 .$$

It follows that for any $N'>1$ and any ν such that $0<\nu<N'-1$,

$$g\left[\frac{N'}{2}-\nu\right]=g\left[\nu+1-\frac{N'}{2}\right] . \tag{26}$$

It follows from Eq. (26) that, if a natural frequency occurs for some value of m given by $N'/2-\nu$ or $\nu+1-N'/2$, resonance must occur for some lower value unless

$$\frac{N'}{2}-\nu=\nu+1-\frac{N'}{2} . \tag{27}$$

Condition (27) is equivalent to

$$N'=2\nu+1 , \tag{28}$$

so that to avoid resonance and still have oscillation, N' must be odd.

Also, on comparing Eq. (26) with Eq. (28), it is found that this can only occur when $m=\frac{1}{2}$, for which value

$$g(m)=g\left[\frac{1}{2}\right]=\left[\frac{N'+1}{2}\right]^2 .$$

Then, the natural frequency oscillation condition Eq. (25) becomes

$$K<(N'+1)\sqrt{2/N'}\sim\sqrt{2N'} . \tag{29}$$

To avoid resonance due to the natural frequency for a larger value of m, the condition

$$K>2\sqrt{g(\tfrac{1}{2})/N'}=2\sqrt{(N'+3)(N'-1)/2N'} \tag{30}$$

must be satisfied.

In summary, the only case in which there is oscillation of the system but no resonance occurs is when

$$\sqrt{2(N'+3)(N'-1)/N'}<K<(N'+1)\sqrt{2/N'} . \tag{31}$$

The case of $N'=2$ provides an example for which the explicit analytical results are reasonably simple. On applying the matrix inverse Eq. (23) to Eq. (20) and inverting the Laplace transforms obtained thereby, the following solutions are obtained:

$$N(1,t)=0, p(1,t)=\frac{4}{8-K^2}e^{-Kt}[1-\cos(\sqrt{8-K^2}\,t+\phi)] ,$$

where

$$\sin\phi=\frac{K\sqrt{8-K^2}}{4}, \quad \cos\phi=\frac{K^2-4}{4} ,$$

$$N(0,t)=\frac{4}{8-K^2}e^{-Kt}[1-\cos\sqrt{8-K^2}\,t] ,$$

$$N(-1,t)=\frac{32}{(8-K^2)^2}e^{-Kt}[1-\cos\sqrt{8-K^2}\,t-\sqrt{(8-K^2)/2}\,t\sin\sqrt{8-K^2}\,t] , \tag{32}$$

$$p(0,t)=\frac{K^2}{8}N(-1,t)+\frac{4}{(8-K^2)^2}e^{-Kt}\left[\sqrt{(8-K^2)/2}\sin\sqrt{8-K^2}\,t-\left[\frac{8-K^2}{2}\right]t\cos\sqrt{8-K^2}\,t+\frac{(8-K^2)^{3/2}}{2}t\sin\sqrt{8-K^2}\,t\right] ,$$

$$p(-1,t)=1-p(0,t)-p(1,t) .$$

The last equation comes from the conservation of probability

$$\sum_m p(m,t)=1 ,$$

which follows identically from Eq. (16) and the assumed initial conditions.

The emitted pulse is given by the sum of the individual photon contributions as in Eq. (17). Figures 6(a)–6(e) show some calculated results from Eq. (32). These results compare well with the calculations obtained using the previous numerical methods of solving (16) for the same input parameters.

V. CONCLUSIONS

The BL mean-field model of superfluorescence due to an array of identical two-level atoms has an important

advantage not shared by other, more recent and more comprehensive models. It deals quantum mechanically with emitters in a crystal lattice, which is the most likely structure for the γ-ray laser.

The model can be applied to a similar structure composed of nuclear emitters that radiate electromagnetic energy at wavelengths smaller than the interatomic spacing. It predicts that superfluorescence in this case is possible only at wavelengths satisfying conditions that are sufficient for Borrmann coupling to occur. However, since the BL model is one dimensional it cannot distinguish among the various Borrmann modes that Trammel, Hutton, and Hannon[18] have discussed in connection with a three-dimensional crystal.

Solutions of the BL model will approach a classical limit some time after superfluorescent radiation begins; however, in some cases the early time during which the classical approximation remains invalid lasts until after the peak of the radiated pulse has emerged. On the other hand, it is found that in some cases the iteration process used to approximate the quantum-mechanical solution, which is required during the early stages of the emission process before the classical limit provides a valid approximation, can, itself, be invalid because of a numerical instability inherent in the process.

ACKNOWLEDGMENTS

The authors would like to thank Professor Joseph Eberly of the University of Rochester for the many helpful discussions which provided much insight into the problem of superfluorescence and for his critical review of the manuscript. This work was supported by the Innovative Science and Technology Directorate of SDIO.

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[1]B. Balko, I. W. Kay, and J. W. Neuberger (unpublished).

[2]Electronic attenuation, competing transitions, inhomogeneous and homogeneous line broadening, and finite pumping times.

[3]Although in general the attenuation of electromagnetic radiation of soft γ rays and x rays in solids is high (typically 10^2-10^3 cm^{-1}) at wavelengths of interest (in the region of 0.1–1 Å) in crystals at special directions the attenuation can be reduced by two or three orders of magnitude. The phenomenon is known as the Borrmann effect.

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[11]Then, e.g., $\alpha = 2\alpha'$ will satisfy Eq. (8).

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[13]R. Bonifacio, P. Schwendimann, and F. Haake, Phys. Rev. 4, 302 (1971).

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[15]R. Glauber and F. Haake, Phys. Lett. 68A, 29 (1978).

[16]K^{-1} is the maximum round trip travel time for the photons in the active volume. As shall be shown in Sec. IV for $K < 2\sqrt{2}$ one obtains ~ pure or single pulse SF because the photons do not spend enough time in the active volume to be reabsorbed and reemitted as multiple pulses.

[17]Ringing will occur because of at least one value of m, namely $m = 0$, when K is of the order of $\sqrt{N}$ or less. To get ringing for all m values $K < 2\sqrt{2}$ is the necessary condition.

[18]Cf. G. T. Trammell, J. T. Hutton, and J. P. Hannon, J. Quant. Spectrosc. Radiat. Transfer 40, 693 (1988).

PAPER No. 2
A FEASIBILITY STUDY OF NUCLEAR SUPERFLUORESCENCE

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Abstract

A theory of nuclear superfluorescence (SF) based on the Haake-Reibold model for the atomic case is presented. Certain modifications of the model make it possible to take into account some effects that are more important in nuclear than in atomic SF: attenuation, competing transitions, finite pumping times, and both homogeneous and inhomogeneous line broadening. Results of some explicit calculations illustrate the influence of these effects on the radiated pulse. Similar calculations are used to examine the feasibility of observing SF using the 58.6 keV transition in ^{60}Co.

I. Introduction

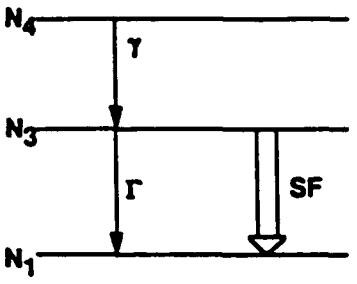
Superfluorescence (SF) (Refs. 1-4) is an example of the spontaneous, cooperative emission of coherent radiation by a collection of identical molecules, atoms or nuclei. Intense, directed pulses of duration much shorter than the spontaneous emission (SE) lifetime of an individual radiator characterize the radiation, which has been observed experimentally for atoms and molecules (Ref. 2), e.g., in CH_3F, HF, Na, Ca, Tl, KCl, Sr, and Li. Existing theoretical models (Ref. 2) have successfully explained the observations.

However, thus far no one has reported an observation of nuclear SF, nor have the theoretical models used to analyze the corresponding nuclear phenomena been adequate. This is unfortunate since SF (Refs. 5, 6) will probably be the most important emission process in a γ-ray laser operation. The emission time of a cooperative pulse is inversely proportional to the number of cooperating emitters, and is therefore likely to be much shorter than the natural nuclear lifetime. Also, the peak intensity of the radiated pulse will be proportional to the square of the number of cooperating emitters and therefore much larger than the intensity of the incoherent SE, which is just proportional to the number of emitters.

This paper takes into account certain experimentally achievable parameters that are more important for nuclear than for atomic SF. Its theoretical treatment is based on the Maxwell-Bloch equations introduced by Haake and Reibold (Ref. 7) to treat superfluorescent phenomena in multi-level systems, but modified here to include most of the important nuclear effects. We apply the theory to a real nucleus, ^{60}Co, created by thermal neutron irradiation of ^{59}Co, and show under what conditions nuclear SF is possible in this system.

II. The Haake-Reibold Model for Nuclear Superfluorescence

Fig. 1 depicts the Haake-Reibold model for SF, including some parameters that cannot be neglected in the nuclear case. As indicated in the diagram on the left, level 4 decays at the rate γ in a pumping process that populates level 3. The inverted population of level 3 then decays with the rate Γ to the ground state at level 1, collectively emitting SF radiation that is proportional to the square of the population. Appearing on the right, the Maxwell-Bloch equations govern the relation between the populations N_4, N_3, N_1 of the three levels, the electric field $E^{\pm}$,the collective polarization $R^{\pm}$, and the source term $\xi^{\pm}$, all of which are functions of distance and time.[1] In general, the electric field satisfying the equations is a pulse propagating from left to right.



$$\frac{\partial N_4}{\partial t} = -\gamma N_4$$

$$\frac{\partial N_3}{\partial t} = -\left(E^{+}R^{+}+E^{-}R^{-}\right) - \Gamma N_3 + \gamma N_4$$

$$\frac{\partial N_1}{\partial t} = +\left(E^{+}R^{+}+E^{-}R^{-}\right) + \Gamma N_3$$

$$\frac{\partial R^{\pm}}{\partial t} = (N_3 - N_1)\,E^{\mp} - \frac{1}{2}(\Gamma + \Gamma_{\phi})R^{\pm} + \xi^{\pm}$$

$$\frac{\partial E^{\pm}}{\partial x} = g(t)R^{\mp} - \frac{1}{2}\mu E^{\pm},$$

The unit of time is

$$\tau_{SF} = \frac{8\pi\tau_0}{3\lambda^2\rho l},$$

where l is the cavity length, λ is the wavelength of the emitted photon, τ_0 is the natural radiative lifetime, and ρ is the density of cooperating nuclei.

Figure 1 The Haake-Reibold Model

In the Maxwell-Bloch equations t represents retarded time, so that the last equation implies an electric field propagating from left to right. Also, the source term $\xi^{\pm}$ represents noise due to fluctuations of the vacuum and is therefore a stochastic quantity, the properties of which Ref. 8 derives using a quantum electrodynamic argument.

---

[1]These quantities are quantum mechanical operators, but because the source term becomes negligible after a short time and the other operators all commute with each other, they can be treated as c-numbers and identified with their expected values.

The SF model depicted in Fig. 1 includes the following phenomena, all of which are important for the nuclear case: electromagnetic attenuation, natural decay, competing transitions, homogeneous and inhomogeneous line broadening, pumping rate, and relaxation. Various parameters appearing in the Maxwell-Bloch equations account for these phenomena and determine their ultimate effect on the system radiation.

As indicated by the first equation, the reciprocal of γ is the effective pumping time. The form of the second equation shows that Γ is the natural decay rate. A quantity μ in the last equation obviously causes attenuation of the propagating electric field.

The homogeneous line broadening mechanism differs from that of the inhomogeneous broadening. The fourth equation indicates that the quantity Γ_ϕ, which represents dephasing associated with homogeneous line broadening, simply adds to the effect of the natural decay. However, the function g(t) in the last equation is responsible for taking into account inhomogeneous line broadening. The form of g(t) depends on the statistical distribution of frequencies ω_j associated with random dephasing factors contributed at a particular location by photons emitted at other positions x_j along the active region. If the distribution is Lorentzian g(t) has the form

$$g(t) = g_0 e^{-\frac{\Gamma_\phi t}{2}}.$$

The quantity g_0 is the coupling constant relating the electric field to the polarization.

III. Predictions of the Modified Haake-Reibold Model

Using a Monte Carlo procedure to simulate the stochastic source ξ^*, numerical solutions of the Maxwell-Bloch equations in Fig. 1 lead to the curves given in Figs. 2-4, which illustrate the effect of attenuation and line broadening on the radiated SF pulse. Fig. 2 shows how the peak intensity and the delay time of the peak vary as functions of μ for a cooperation number N equal to 10^3. Fig. 3 illustrates the effect of homogeneous and inhomogeneous line broadening on the SF pulse shape, using curves representing intensity as a function of time for this purpose. Fig. 4 shows the delay time as a function of homogeneous and inhomogeneous broadening. The figures use a parameter a equal to the ratio of Γ_ϕ to the natural decay rate Γ and a parameter b equal to the ratio of Γ_ϕ to Γ as convenient measures of line broadening.

Ref. 9, reporting on observations of combined SF and amplified spontaneous emission (ASE) produced by a $KCl:O_2^-$ atomic system, gives experimental curves showing the effect of line broadening on the delay time of emitted pulses of both types. Calculations based on the Haake-Reibold model used to predict SF and ASE agree quite well with the Ref. 9 results when reasonable values are chosen for parameters that could not be measured or otherwise determined.

Unfortunately, no one has yet reported a similar experiment involving nuclear rather than atomic emissions. However, Ref. 10 gives a list of candidates to replace $KCl:O_2^-$ in such an experiment, one of which is ^{60}Co prepared in the excited isomeric state by thermal neutron pumping of ^{59}Co.

Aside from this pumping mechanism, other advantages of ^{60}Co are: (1) its lifetime, 906

sec., which would allow long pumping times for population inversion and therefore would involve achievable thermal neutron fluxes; (2) the fact that its natural lifetime is, nevertheless, relatively short; (3) its relatively good Mössbauer effect; (4) its low losses due to such factors as internal conversion and electronic attenuation.

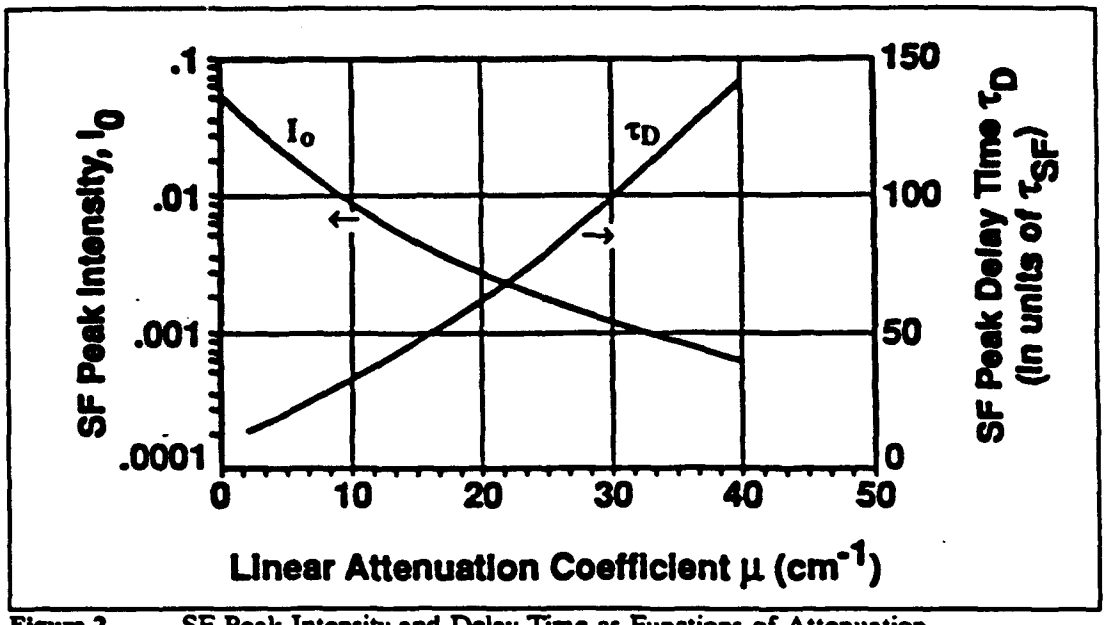


Figure 2 SF Peak Intensity and Delay Time as Functions of Attenuation

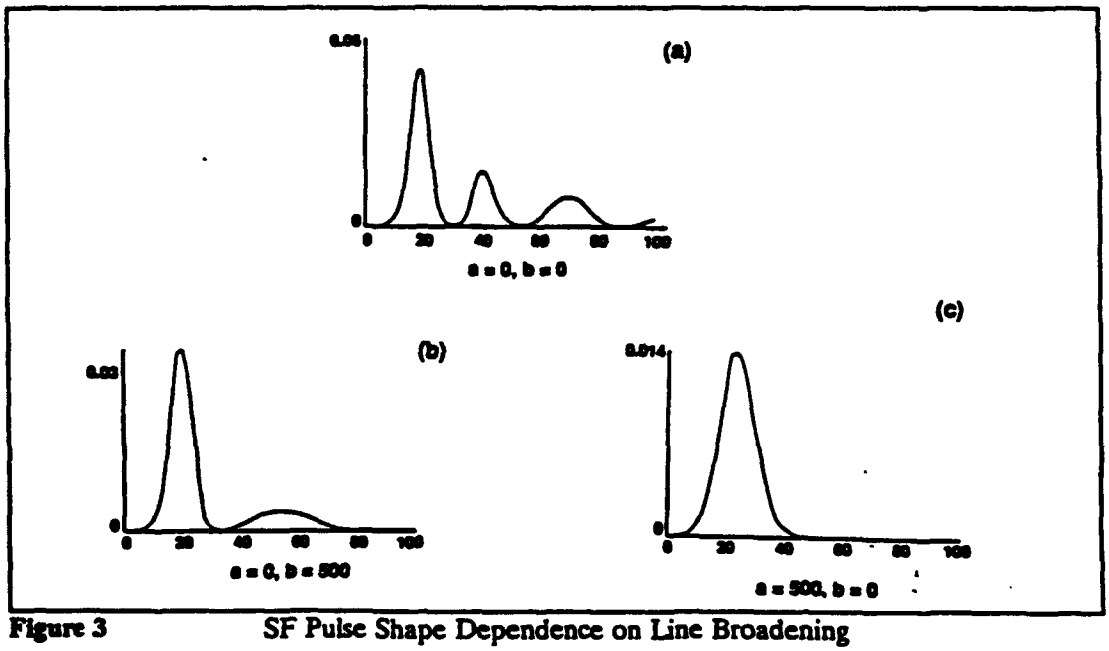


Figure 3 SF Pulse Shape Dependence on Line Broadening

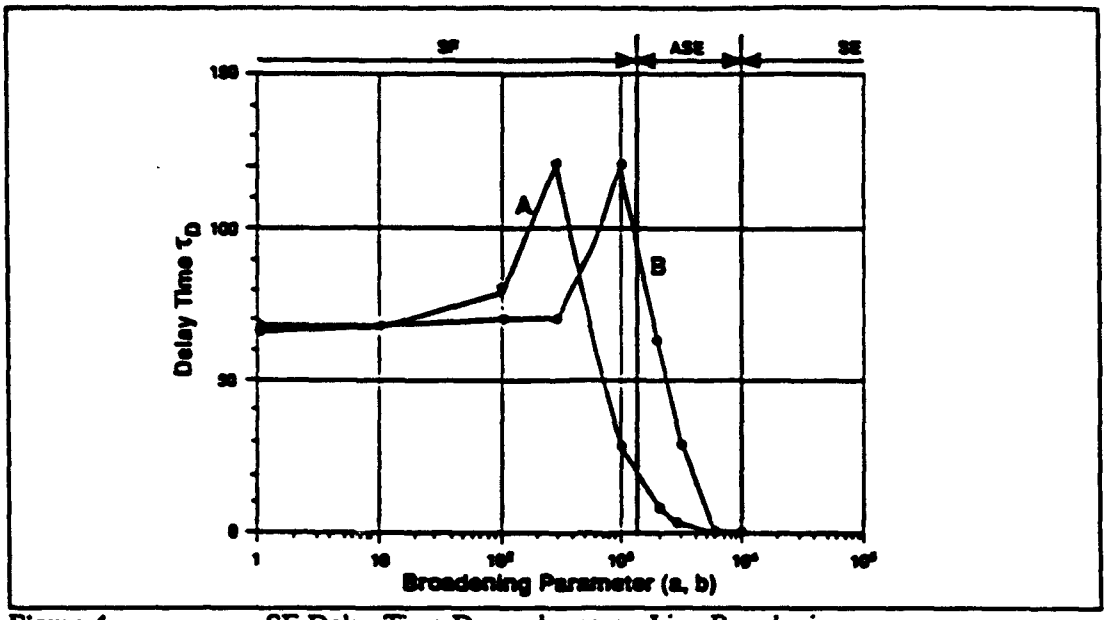


Figure 4 SF Delay Time Dependence on Line Broadening

Table 1 gives the characteristic atomic and nuclear parameters associated with a single ^{60}Co nucleus. It omits inhomogeneous broadening since that phenomenon involves multiple nuclei. In fact, the only estimates of inhomogeneous broadening in connection with a long lived, Mössbauer effect isotope that are available in the literature are for ^{107}Ag (Ref. 11) and ^{109}Ag (Refs. 12-14) and are based on self-attenuation measurements. The earliest estimates of the inhomogeneous broadening parameter a were of the order 10^5, but more recent ones (Refs. 13, 14) are much smaller, ranging from 30 to 200.

Table 1 gives 12 cm^{-1} for the electronic attenuation μ. However, exploiting the Borrmann effect in a single crystal can reduce that value to ~0.01 cm^{-1}.

In a generic example, Fig. 5 compares the ^{60}Co SF intensity as a function of time, calculated by means of the Haake-Reibold model, with a similar calculation of the SE. It shows the number of photons emitted per sec per nucleus for both cases separately and combined. Whether the SF is detectible in an experiment will depend on whether the disturbance it causes in the combined SF and SE is measurable.

If the fluorescent material is a single, acicular shaped crystal the SF will radiate in an endfire mode in a narrow beam, with a high gain relative to an isotropic radiation pattern characteristic of SE, due to natural decay. If the diameter d of the active region is related to its length l and the wavelength λ by the Fresnel condition

$$d = \sqrt{\lambda l}$$

for a minimum beamwidth β, i.e., angular divergence due to diffraction, the beamwidth is given

by

$$\beta = \frac{\lambda}{d}.$$

Table 1

Characteristic Parameters for ^{60}Co

Parameter	Value	Symbol
Transition energy	58.6 keV	E_θ
Wavelength on resonance	0.2Å (2×10^{-8} cm)	λ
Natural lifetime	628 sec	τ_θ
Recoilless fraction	0.304	f
Linear attenuation coefficient	12 cm^{-1}	μ
Interval conversion coefficient	48.3	α
Particle density (in solid form)	8.97×10^{22} cm^{-3}	ρ
Thermal neutron Cross-section $^{59}Co+n \rightarrow {}^{60}Co$	20 barns	σ_a

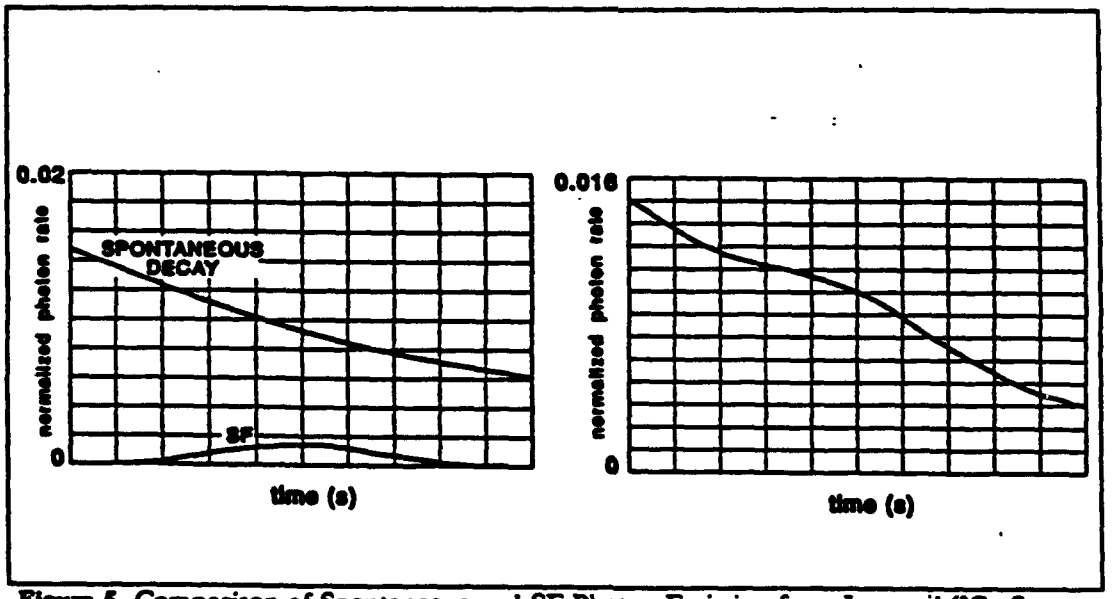


Figure 5 Comparison of Spontaneous and SF Photon Emission from Inverted ^{60}Co System

If the SF beamwidth is 5×10^{-5} rad, which corresponds to a solid angle of $6.25 \times 10^{-10}\pi$ sterad, the gain relative to an isotropic polycrystalline radiator will be $\sim 6.4 \times 10^{9}$. Neglecting inhomogeneous broadening and attenuation, Table 2 gives the normalized photon emission rate for both the case of a polycrystalline sample, labelled Experiment A, and a single crystal, labelled Experiment B, for different values of the thermal neutron flux used to pump the ^{59}Co nucleus.

An examination of Table 2 shows that in the B experiment for a thermal neutron flux of 2.1×10^{17} cm^{-2}sec^{-1} the pumping rate γ creates an SF photon emission rate in the direction of maximum radiation 100 times higher than that of the coexisting SE. This implies that a thermal neutron flux not much less than 2×10^{17} is needed to observe SF in the 58.6 keV transition of ^{60}Co, when both the inhomogeneous broadening effect is small and the Borrmann effect is strong.

Table 2

Predictions for ^{60}Co Experiments

		Total Count Ratio SF/SE	
		Experiment A	Experiment B
		Polycrystal	Single crystal acicular rod
Case 1	$J = 2.1 \times 10^{23}$cm^{-2}sec^{-1} $a = 0$ $b = 0$ $\mu = 0$	2.9	1.9×10^{10}
Case 2	$J = 2.1 \times 10^{23}$cm^{-2}sec^{-1} $a = 50$ $b = 0$ $\mu = 0$	10^{-4}	6.4×10^{5}
Case 3	$J = 2.1 \times 10^{23}$cm^{-2}sec^{-1} $a = 20$ $b = 0$ $\mu = 12$ cm^{-1}	8.6×10^{-6}	5.5×10^{4}
Case 4	$J = 2.1 \times 10^{17}$cm^{-2}sec^{-1} $a = 0$ $b = 0$ $\mu = 0$	1.6×10^{-6}	100

IV. Conclusions

The suggested experiment with ^{60}Co as a source of nuclear SF appears to be feasible. The thermal neutron pumping flux required to prepare the isomer in its inverted state is of the order of 10^{17}neutrons cm^{-2}sec^{-1}, which is within reach of existing technology[2]. Analyses similar to that presented in this paper applied to other candidates, perhaps involving other pumping methods, may uncover even more promising nuclear SF sources.

Acknowledgment

This research is supported by SDIO/IST under contract MDA 903-89-C-003.

[2]Report of the Neutron Source Committee, Oak Ridge National Laboratory, May 20, 1976 (unpublished).

PAPER No. 3

A FEASIBILITY STUDY OF NUCLEAR SUPERFLUORESCENCE

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A FEASIBILITY STUDY OF NUCLEAR SUPERFLUORESCENCE

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I. Introduction

Superfluorescence (SF) (Refs. 1-3) is an example of the spontaneous, cooperative emission of coherent radiation by a collection of identical molecules, atoms or nuclei. Intense, directed pulses of duration much shorter than those due to the spontaneous emission (SE) or amplified spontaneous emission (ASE) pulses characterize the radiation. The emission time of a cooperative pulse is inversely proportional to the number of cooperating emitters, and is therefore likely to be much shorter than the natural lifetime. Also, the peak intensity of the radiated pulse is proportional to the square of the number of cooperating emitters and therefore much larger than either the intensity of the incoherent SE, which is just proportional to the number of emitters, or the intensity of ASE. Although the kinetics of stimulated γ-radiation has been discussed in the literature (Ref. 4, 5, 6), nuclear SF, which may be the most important process in a γ-ray laser operation, has not been adequately treated.

SF has been observed for atoms and molecules (Ref. 2). Existing theoretical models (Ref. 2) have successfully explained the observations. It has also been observed experimentally in atomic and molecular systems that as the dephasing rate between the emitters increases, the SF pulse shape changes into a characteristic amplified spontaneous emission (ASE) pulse shape: the peak shifts to longer times, the pulse width increases and intensity becomes proportional to the number of emitters. However, thus far no one has reported an observation of nuclear SF, nor have the theoretical models used to analyze the corresponding nuclear phenomena been adequate for predicting whether nuclear SF is possible.

In this paper we present a theory of nuclear superfluorescence (SF) based on the Haake-Reibold model for the atomic case (Ref. 7). Certain modifications of the model make it possible to take into account some effects that are more important in nuclear than in atomic SF: attenuation, competing transitions, finite pumping times, and both homogeneous and inhomogeneous line broadening. Results of some explicit calculations illustrate the influence of these effects on the radiated pulse. An extension of this theory can be used to model ASE and the transition from SF to ASE as a function of the dephasing rate (homogeneous and inhomogeneous broadening). To check the validity of our model we compared calculated pulse shapes with published experimental results obtained with $KCl:O_2$ (Ref. 8) in the SF and ASE regimes and the transition region. As a final example, we apply the theory to a real nucleus, ^{60}Co, created by thermal neutron irradiation of ^{59}Co, and show under what conditions nuclear SF is possible in this system.

II. The Haake-Reibold Model for Nuclear Superfluorescence: Verification and Prediction

Fig. 1 displays the Haake-Reibold model for SF, including some parameters that cannot be neglected in the nuclear case. The diagram on the left shows the level scheme and the transitions assumed in this paper. The Maxwell-Bloch equations on the right govern the relation between the populations N_4, N_3, N_1 of the three levels, the electric field $E^{\pm}$, the collective polarization $R^{\pm}$,, and the source term $\xi^{\pm}$, all of which are functions of distance and time. The real time and space coordinates are normalized to the superfluorescence time and cavity length, respectively. The superfluorescence time is given by

$$\tau_{SF} = 8\,\pi\tau_o/3\lambda^2\rho l\ ;$$

l is the cavity length, λ is the wavelength of the emitted photon, τ is the natural radiative lifetime, and ρ is the inversion density of cooperating nuclei.

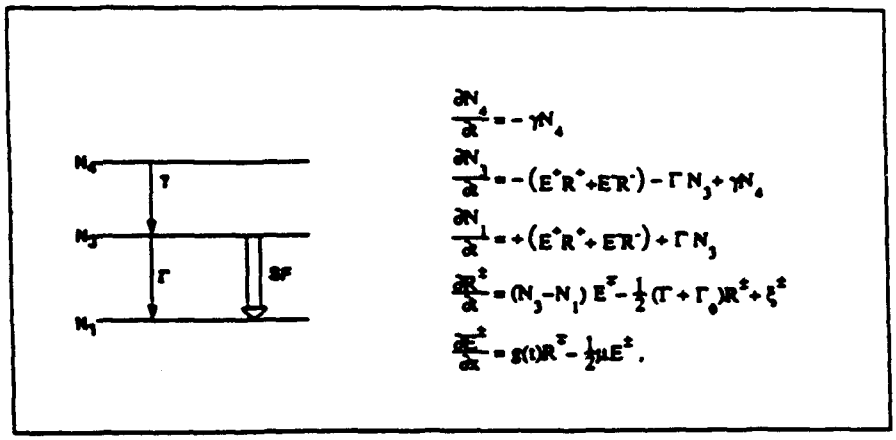

Figure 1. The Haake-Reibold Model

In the equations τ represents retarded time, the source term $\xi^\pm$ (a stochastic quantity) represents noise due to fluctuations of the vacuum the properties of which Ref. 9 derives using a quantum electrodynamic argument. The SF model, as indicated in Fig. 1, includes electromagnetic attenuation, natural decay, competing transitions, homogeneous and inhomogeneous line broadening, pumping rate, and relaxation. The reciprocal of γ is the effective pumping time, Γ is the natural decay rate and μ causes attenuation of the propagating electric field. The homogeneous line broadening mechanism differs from that of the inhomogeneous broadening. The quantity Γ_ϕ, which represents dephasing associated with homogeneous line broadening, simply adds to the effect of the natural decay while the function g(t) takes into account inhomogeneous line broadening. The form of g(t) depends on the statistical distribution of frequencies ω_j associated with random dephasing factors contributed at a particular location by photons emitted at other positions x_j along the active region. If the distribution is Lorentzian g(t) has the form

$$g(t) = g_o \exp(-\Gamma_\theta t/2)$$

g_o is the coupling constant relating the electric field to the polarization.

Using a Monte Carlo procedure to simulate the stochastic source $\xi^\pm$, numerical solutions of the Maxwell-Bloch equations in Fig. 1 demonstrate both the attenuation μ and the line broadening Γ_θ or Γ_ϕ. Inhomogeneous broadening has a more drastic effect on the line shape than homogeneous broadening for the same amount of broadening.

Ref. 8, reporting on observations of combined SF and amplified spontaneous emission (ASE) produced by a KCl 0_2 atomic system, gives experimental curves showing the effect of line broadening on the delay time of emitted pulses of both types. We performed calculations based on the Haake-Reibold to compare with the experimental results. Reasonable values were chosen for parameters that could not be measured or otherwise determined.

In the Haake-Reibold (Fig. 7) model the stochastic source term is proportional to the pumping rate γ and decays exponentially with the rate γ. This source, $\xi^\pm$ generates SF pulses

agreeing with experimental results in the SF regime but not in the ASE regime. In the ASE regime a stochastic source with γ replaced by Γ, the spontaneous lifetime of the excited state, provides agreement with experiment. Alternatively an incoming pulse at the left end of the active region with an amplitude decaying at the rate Γ can be used to model ASE.

Figure.2 shows the comparison of the theoretical calculations with the experimental data for five different temperatures (Ref. 8). The result at 10K which we assumed to be due to pure SF was used to determine the size of the active volume or cooperation number N and the SF time τ_{SF}. The rest of the calculated results were obtained using N-10^9, an inhomogeneous broadening of 400 times the natural line width and homogeneous broadening determined by the respective temperatures as shown in the figure.

In Figure 2 the experimental pulses shown are reproduced from Ref. 8. The solid curves give the calculated results obtained with τ_{SF} = 1.82 ps. The dashed curves represent calculated results with τ_{SF} doubled.

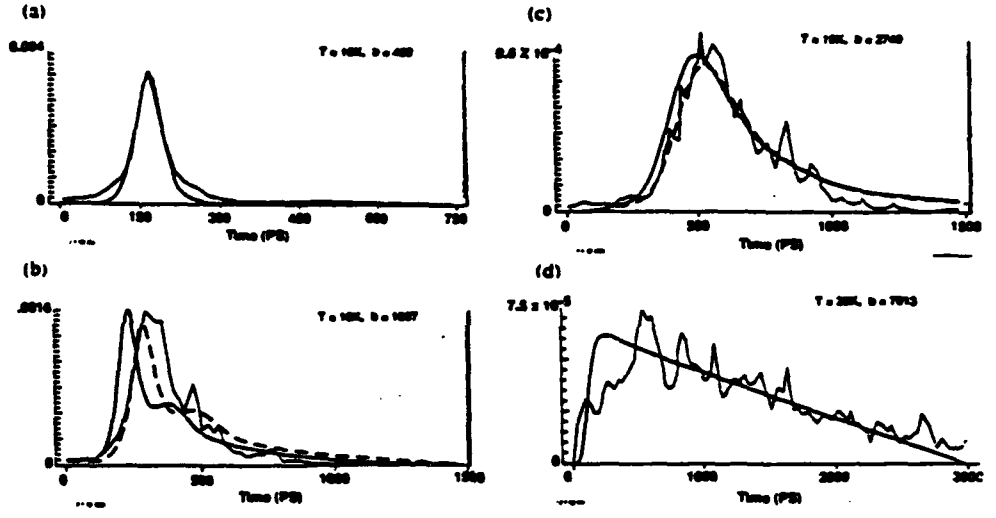

Figure 2 Experimental and Theoretical SF and ASE pulse shapes from KCl:O^-_2

Calculations based on the Haake-Reibold model used to predict SF and ASE agree quite well with the Ref. 8 results when reasonable values are chosen for parameters that could not be measured or otherwise determined.

Unfortunately, no one has yet reported a similar experiment involving nuclear rather than atomic emissions. However, Ref. 10 gives a list of candidates to replace KCl:O^-_2 in such an experiment, one of which is ^{60}Co prepared in the excited isomeric state by thermal neutron pumping of ^{59}Co.

Aside from this pumping mechanism, other advantages of ^{60}Co are: (1) its lifetime, 906 sec., which would allow long pumping times for population inversion and therefore would involve achievable thermal neutron fluxes; (2) the fact that its natural lifetime is, nevertheless, relatively short and thus its coupling with the electromagnetic field relatively strong; (3) its relatively good Mössbauer effect; (4) its low losses due to such factors as internal conversion and electronic attenuation.

Using characteristic parameters for ^{60}Co and the thermal neutron cross-section for preparing ^{60}Co from ^{59}Co we calculated the SF pulse intensity for different values of inhomogeneous and homogeneous broadening and different thermal neutron fluxes. Assuming an angular shape for the active material we compared the SF intensity in an end fire mode with the natural decay component along the axis. We found that for a thermal neutron flux of 2.1×10^{17} $cm^{-2}sec^{-1}$ the pumping rate γ creates an SF photon emission rate in the direction of maximum radiation 100 times higher than that of the coexisting SE. This implies that a thermal neutron flux can be used to observe SF in the 58.6 keV transition of ^{60}Co, when both the inhomogeneous broadening effect is small and the Borrmann effect which reduces the attenuation in certain directions in a crystal, is strong. Reactors providing such thermal neutron fluxes have been designed and may become available (Ref. 9, 10).

III. Conclusions

The Haake-Reibold model for SF has been generalized to treat nuclear SF and ASE. A comparison of theoretical results with experimental pulses obtained from $KCl:0_2$ gives good agreement for atomic SF, ASE and the transition region. This agreement with experiment justifies some confidence in the model for predicting the conditions required for the occurrence of nuclear SF.

The suggested experiment with ^{60}Co as a source of nuclear SF appears to be feasible. The thermal neutron pumping flux required to prepare the isomer in its inverted state is of the order of 10^{17} neutrons $cm^{-2}sec^{-1}$, which is within reach of existing technology. Analyses similar to that presented in this paper applied to other candidates, perhaps involving other pumping methods, may uncover even more promising nuclear SF sources.

Acknowledgement

This research is supported by SDIO/IST under contract MDA 903-89-C-003.

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PAPER No. 4

COMMENT ON "NUCLEAR RESONANT ABSORPTION IN LONG-LIVED ISOMERIC TRANSITIONS"

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Comments

Comments are short papers which criticize or correct papers of other authors previously published in the Physical Review. *Each Comment should state clearly to which paper it refers and must be accompanied by a brief abstract. The same publication schedule as for regular articles is followed, and page proofs are sent to authors.*

Comment on "Nuclear resonant absorption in long-lived isomeric transitions"

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(Received 27 October 1992)

In a recent paper [R. Coussement *et al.*, Phys. Rev. B 45, 9755 (1992)] it was suggested that for a long-lived state that can undergo a Mössbauer transition, if the homogeneously broadened width, rather than the natural width, exceeds the solid-state-induced inhomogeneous width, it should be possible to observe resonant emission and absorption. On the other hand, it is expected that the probability of absorption from a spectral distribution of total width Γ should be proportional to Γ_γ/Γ, where Γ_γ is the radiation width of the absorbing nucleus. Generally Γ is expected to be larger than Γ_γ because of the possibilities of internal conversion, transitions between magnetic sublevels, etc., all of which contribute to homogeneous broadening and should reduce the Mössbauer effect. The present Comment addresses questions of principle, mathematical assumptions, and other aspects of the work of Coussement *et al.* and demonstrates that a correct statistical treatment recovers the conventional more stringent requirement on the broadening.

Coussement *et al.*[1] have argued that the usual criterion for the observation of Mössbauer self-absorption is too pessimistic: "It is merely sufficient for the inhomogeneous width (or detuning distribution) to be small compared with the width of the Fourier spectrum of the fluctuations, rather than with the natural linewidth." However, this conclusion is in error, due primarily to an inconsistent treatment of the random factors in the wave functions. While we have a number of other problems with the details of their treatment, they are incidental to our central point. We shall therefore accept their initial assumptions and demonstrate that a correct statistical treatment recovers the conventional, more stringent requirement on the broadening. In the second part of the Comment we demonstrate the distinction between the overlap of spectral lines and the resonant absorption of a spectral line and show that contrary to the contention of Ref. 1 complete overlap of lines does not guarantee a strong Mössbauer effect.

According to Ref. 1, Eq. (8), the probability that a transition has taken place is proportional to the following integral. (The amplitude factor is omitted for simplicity.)

$$P^T(\delta\omega)\equiv\left|\int_0^T dt\, f(t)g^*(t)e^{-\Gamma_a t-i\delta\omega t}\right|^2, \tag{1}$$

where $\delta\omega=\omega_0-\omega_0'$ is the energy difference in the absence of the random fluctuation factors $f(t)$ and $g(t)$ for the emitter and absorber, respectively, and Γ_a is the natural linewidth of the Mössbauer transition. The distinction between the natural linewidth Γ_a, the total linewidth Γ, and the radiative linewidth Γ_γ is not made in Ref. 1 and thus the physical result we derive here is notationally forbidden in Ref. 1. In Ref. 1 the energy difference $\delta\omega$ is described as due to the detuning but, as will be discussed below, a more general interpretation is possible. Reference 1 then considers an average rate defined by

$$R^T(\delta\omega)\equiv\frac{P^{T+\Delta T}(\delta\omega)-P^T(\delta\omega)}{\Delta T}, \tag{2}$$

where ΔT is long compared to the fluctuation time scale of $f(t)$ and $g(t)$ but short compared to the nuclear lifetime. Under these conditions the temporal average implicit in the ratio of the above expression should be equivalent to computing the ensemble average of $P^T(\delta\omega)$. If the rate for intermediate times is then desired, the rate is just given by the derivative of $P^T(\delta\omega)$. We will therefore concentrate on a correct evaluation of the probability $P^T(\delta\omega)$. Rewrite $P^T(\delta\omega)$ as

$$P^T(\delta\omega)=\int_0^T dt'\int_0^T dt\, f(t)f^*(t')g(t')g^*(t)\times e^{-\Gamma_a(t+t')-i\delta\omega(t-t')}. \tag{3}$$

Taking the ensemble average over the random fields f and g and assuming them to be independent and stationary (as in Ref. 1) one has

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$$P^T(\delta\omega)=\int_0^T dt'\int_0^T dt[|\bar{f}|^2+C_f(t-t')]\times[|\bar{g}|^2+C_g^*(t-t')]\times e^{-\Gamma_n(t'+t)-i\delta\omega(t-t')}, \quad (4)$$

where $\bar{f}$ and $\bar{g}$ are the ensemble averages of $f(t)$ and $g(t)$ [$\bar{f}=E(f)$], $\bar{g}=E(g)$, where $E(\ )$ denotes the expectation value over the random perturbations)[2] and C_f and C_g are the two-point correlation functions.[3]

Denoting the Fourier transforms of C_f and C_g by $F(\omega)$ and $G(\omega)$ and defining $Q(\omega)$ to be the convolution of $F(\omega)$ and $G(-\omega)$, Eq. (4) can be written as

$$P^T(\delta\omega)=\int\frac{d\omega}{2\pi}W(\omega)\frac{1-2\cos[(\delta\omega+\omega)T]e^{-\Gamma_n T}+e^{-2\Gamma_n T}}{\Gamma_n^2+(\delta\omega+\omega)^2}, \quad (5a)$$

with

$$W(\omega)=Q(\omega)+|\bar{f}|^2G(-\omega)+|\bar{g}|^2F(\omega)+2\pi\delta(\omega)|\bar{f}|^2|\bar{g}|^2. \quad (5b)$$

Thus, the resulting probability is a weighted sum of standard terms, which reduce to Lorentzians for large T, the first term persisting even if the average values of $f(t)$ and $g(t)$ are zero.[4]

Equation (5a) can be simply evaluated for distributions of interest. For example, if we assume that the average values of $f(t)$ and $g(t)$ vanish and the autocorrelation functions are exponential with time constant $1/\Delta$, then for large times,

$$P(\delta\omega)=\frac{1}{\Gamma_n}\frac{\Gamma_n+\Delta}{(\delta\omega)^2+(\Gamma_n+\Delta)^2}. \quad (6)$$

When the broadening due to the random fluctuations, Δ, is small compared to the basic linewidth, the unperturbed Lorentzian line shape is recovered. On the other hand, for large Δ, the probability at the peak $\delta\omega=0$ is inversely proportional to $\Gamma_n\Delta$.

A similar result is obtained if a Gaussian autocorrelation function is assumed. In this case, we have[5]

$$P(\delta\omega)=\sqrt{\pi/2}\frac{1}{\Gamma_n\Delta}\mathrm{Re}[\exp(z)^2\,\mathrm{erfc}(z)], \quad (7a)$$

$$z=\frac{\Gamma_n+i\delta\omega}{\sqrt{2}\Delta}, \quad (7b)$$

where Δ is the standard deviation of the Gaussian distribution of $Q(\omega)$. The behavior of this expression is more complicated than that of Eq. (6) but it has the same limits for large and small Δ. Similar expressions can be obtained for finite values of T.

For both the exponential and Gaussian distributions, the price paid for the homogeneous broadening is that the absorption rate on resonance is decreased from the unbroadened case, by the same factor as the increase in width, with the probability inversely proportional to $\Gamma_n\Delta$. In contrast, the probability given in Ref. 1 would be inversely proportional to Γ_n^2.

In Ref. 1, $\delta\omega$ is described as the detuning of the emitting and absorbing states. It refers to one pair of an absorber and emitter with slightly different resonance energies. The interaction between the nucleus and its environment is different at different lattice sites in the crystal. This provides a distribution of resonant energies and is the source of inhomogeneous broadening. The final result for the probability of absorption would require an integration over $\delta\omega$ to obtain the effect of inhomogeneous broadening. In this case, Δ represents a random broadening separate from the fundamental inhomogeneous broadening. The distribution of the inhomogeneous broadening detuning terms is not discussed in Ref. 1. However, it is easy to see within the formalism given here that if the distribution of inhomogeneously broadened states is of the same form as the correlation function used for $f(t)$ and $g(t)$, then the result is of the same form as given above with an increased value of the broadening term:

$$\Delta_{\text{total}}=\Delta_f+\Delta_{\text{inhomo}}, \quad (8)$$

where Δ_f is the homogeneous broadening due to the fluctuation $f(t)$. The value of $\delta\omega$ [Eqs. (6) and (7)] after such an integration would be any remaining energy difference between the mean energies of the inhomogeneously broadened states, for example, that given by any Doppler shift between emitter and absorber.

On the other hand, one can use the formalism given above to represent the inhomogeneously broadened terms directly. For a set of fixed states with energies shifted by ε_k, the ensemble average has the interpretation of a sum over the possible k states of the emitter and absorber and

$$C_f(t-t')=\sum_k p(\varepsilon_k)e^{-i\varepsilon_k(t-t')} \quad (9)$$

representing a discrete sum of individual unbroadened lines. Note that this form of the correlation function is not necessarily real. Assuming the forest of lines to be essentially dense, using either an exponential or Gaussian distribution for $p(\varepsilon_k)$ and replacing the sum by an integral reproduces the results given in Eqs. (6) and (7) with Δ representing the inhomogeneous width and $\delta\omega$ being the difference between the means of the emitting and absorbing states (typically, the Doppler shift).

These additional considerations do not support the Ref. 1 conclusion that homogeneous broadening due to relaxation increases the recoilless self-absorption in the $^{109}Ag_{47}$ experiment, even though overlap of lines may be achieved. Overlap of lines is not a sufficient condition for a strong resonant absorption effect as the authors of Ref. 1 claim. A vanishing effect might be expected on purely physical grounds in the limit of large homogeneous broadening even as the overlap between the emission line and the absorption line increases.

An illustrative model that distinguishes between the overlap of spectral lines and the probability of emission followed by absorption consists of two Lorentzian lines with a common (unperturbed) resonance energy E_0 and common total width Γ, which are displaced from each

other by 2δ. The overlap of these two lines is given by

$$A(\Gamma,\delta)=\int_{-\infty}^{E_0} dE\, I_2(E)+\int_{E_0}^{\infty} dE\, I_1(E)\,, \tag{10a}$$

where

$$I_{1,2}(E)=\frac{(\Gamma/2\pi)}{(E-E_0\mp\delta)^2+(\Gamma/2)^2}\,. \tag{10b}$$

Performing the integration yields

$$A(\Gamma,\delta)=1-(2/\pi)\tan^{-1}(2\delta/\Gamma)\,, \tag{11}$$

in the limit $\Gamma\to\infty$, the limit of large homogeneous broadening, the area $A(\Gamma,\delta)\to 1$ and complete overlap obtains. This result, however, does not imply that there is a strong resonance effect.

The resonance effect for a nuclear emission process followed by an absorption process is given by

$$R(\Gamma_\gamma,\Gamma,\delta)=\int_{-\infty}^{\infty} dE \frac{I_0(\Gamma/2\pi)}{(E-E_0+\delta)^2+(\Gamma/2)^2} \times\frac{\sigma_0\Gamma_\gamma\Gamma/4}{(E-E_0-\delta)^2+(\Gamma/2)^2}\,, \tag{12}$$

where the resonance energies are displaced from each other by 2δ. The first Lorentzian represents the beam profile and is normalized to unity so that the beam intensity is given by I_0. The second Lorentzian represents the scattering or absorption of a photon with a radiation width for the process Γ_γ, a total width Γ, and σ_0 is the on-resonance cross section $\lambda^2(2I_e+1)/[2\pi(2I_g+1)(1+\alpha)]$, with I_e (I_g) the excited- (ground-) state spin, the internal conversion coefficient is α, and λ is the nominal wavelength of the radiation.

Performing the integration of Eq. (12) yields

$$R(\Gamma_\gamma,\Gamma,\delta)=\frac{I_0\sigma_0}{2}\left|\frac{\Gamma_\gamma}{\Gamma}\right|\frac{1}{1+(2\delta/\Gamma)^2}\,. \tag{13}$$

Increasing the detuning, δ, decreases the resonance effect. For a large detuning, $\delta\gg\Gamma$, and for minimal homogeneous broadening $\Gamma=\Gamma_\gamma$, and therefore $R(\Gamma_\gamma,\Gamma_\gamma,\delta)\approx\Gamma_\gamma^2/\delta^2\ll 1$. As the homogeneous broadening increases the resonance effect increases, but even at the maximum, $\Gamma=2\delta$, is still small since $R(\Gamma_\gamma,2\delta,\delta)\approx\Gamma_\gamma/\delta\ll 1$. Increasing the homogeneous broadening beyond $\Gamma=2\delta$ decreases the resonance effect unless Γ_γ is identical with Γ, i.e., unless the transition of interest is responsible for the total width and Γ_γ is assumed not to be constant. This is not a realistic assumption, however, since in nuclear systems of interest the total width is usually dominated by internal conversion and other processes while the radiative width is only a small part. Not making a distinction between Γ_γ and Γ in Eq. (13) has the same effect as using the overlap in Eq. (11) to calculate this resonance effect. Alternatively, introducing Γ_γ, for Γ in the numerator of (10b) reduces the overlap in Eq. (11) by the factor Γ_γ/Γ. Since we can hardly change the radiative width Γ_γ of a nuclear transition but we can easily change the total or homogeneous width, it is important to keep these distinctions clear in the analysis.

To model the inhomogeneous broadening the integral over appropriate distribution of the detuning parameter δ in Eq. (13) must be taken. In another paper[5] the result is shown to be the error function of Eq. (7a) multiplied by the appropriate factor. When magnetic sublevels may relax among themselves, increasing homogeneous broadening increases the relaxation rate and Γ but does not alter Γ_γ. In the limit of infinite homogeneous broadening, the total width $\Gamma\to\infty$, and although the last factor on the right in Eq. (13) tends to unity, the ratio $\Gamma_\gamma/\Gamma\to 0$. This result remains valid when the present treatment is generalized.

In another paper,[5] we present a general treatment of the emission and absorption of photons by nuclei with inhomogeneously broadened linewidths. This treatment, based on the results of Boyle and Hall[6] and Heitler[7] is more appropriate for the analysis of Mössbauer experiments with isomeric nuclei.

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[1] R. Coussement, G. S'heeren, M. Van Den Bergh, and P. Boolchand, Phys. Rev. B 45, 9755 (1992).

[2] Note that Ref. 1 has an inconsistency in the treatment of $E(f)$ and $E(g)$. It is implied that the average of f and that the average of the magnitude-squared of $f(t)$ are both unity [$E(f)=1$ and $E(ff^*)=1$]. This cannot hold unless f is identically one. If one were to assume as in Blume and Tjon [Phys. Rev. 165, 446 (1968)] that $f(t)$ and $g(t)$ were random phase functions, i.e., that $f(t)=\exp[i\phi(t)]$, the latter condition would hold identically and one might expect $E(f)=0$. For the purposes of this comment the precise value of $E(f)$ is unimportant.

[3] One expects the correlation functions to be real for random homogeneous broadening but there is no loss in assuming them to be complex; the difference expressed in Eq. (4) corresponds to the difference between emitting and absorbing sites.

[4] If the correlation functions C_f and C_g are real, F, G, and Q are even. In any event, F, G, and Q are always real. The use of $G(-\omega)$ in contrast to $F(\omega)$ reflects in Ref. 3 the effect at emitting and absorbing sites.

[5] B. Balko, L. Cohen, D. A. Sparrow, and J. F. Nicoll (unpublished). See also B. Balko, G. Herling, I. W. Kay, and J. Nicoll (unpublished).

[6] A. J. F. Boyle and H. E. Hall, Rep. Prog. Phys. 25, 44 (1962).

[7] W. Heitler, *The Quantum Theory of Radiation*, 3rd ed. (Oxford University Press, London, 1954).

PAPER No. 4A

REPLY TO "COMMENT ON 'NUCLEAR RESONANT ABSORPTION IN LONG-LIVED ISOMERIC TRANSITIONS'"

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Reply to "Comment on 'Nuclear resonant absorption in long-lived isometric transitions' "

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The physical assumptions made to calculate resonant absorption for the nuclear transitions from long-lived isomeric ($\tau > 1$ ms) states by Coussement *et al.* [Phys. Rev. B 45, 9755 (1992)] differ qualitatively from those made to calculate nuclear resonant absorption for transitions from the conventional short-lived ($\tau < 0.1$ ms) states. It is for this reason that one cannot mathematically deduce the resonant fraction for the case of the long-lived states by mere extrapolation of the formulas from the case of short-lived ones.

In our paper[1] we have claimed that the Mössbauer effect in long-lived nuclear states ($\tau > 1$ s) will be possible if the inhomogeneous width (Γ_{inh}) is less that the homogeneous or relaxation width (Γ_r) rather than the natural width (Γ_n). Our claim in this context concerns specifically (a) long-lived states and (b) with both the source and absorber matrices homogeneously broadened by fluctuations. In Mössbauer spectroscopy with conventional short-lived nuclear states ($\tau < 1$ μs), it is well known that to observe the resonance effect, the inhomogeneous width Γ_{inh} should be less than Γ_n, representing a more stringent requirement, which we clearly accept. We believe that in their Comment, Balko *et al.*[2] argue that our claim of the less stringent requirement, viz., $\Gamma_{inh} < \Gamma_r$, to observe the Mössbauer effect in long-lived states is actually in error due to an inconsistent treatment of random factors in the wave function. Balko *et al.*[2] claim that if the algebra is done right, one recovers the more stringent requirement, viz., $\Gamma_{inh} < \Gamma_n$ to observe the Mössbauer effect in long-lived isotopes as well. We disagree with this conclusion. We would like to address the issue in two parts, at first conceptually and then mathematically.

Conceptually, the requirement to observe the Mössbauer effect in long-lived states differs qualitatively from the case of short-lived ones, because of the widely different temporal behavior of the resonant absorption process in the two cases. In the long-lived case ($\tau \sim$ seconds), as we indicated in our paper, the interaction of the electromagnetic (EM) field with nuclei ($\hbar/\tau \simeq 10^{-17}$eV) becomes much weaker than the interaction of nuclei with its environment ($\hbar/\tau_r \simeq 10^{-14}$ eV) in a crystalline solid. This implies that during the long time that nuclear resonant absorption occurs, nuclei in a crystalline solid will have ample opportunity to exchange energy with their environment, and one expects the detuning width or resonance width to be homogeneously broadened by a characteristic relaxation time τ_r (of order of few ms) of the ensemble. The absorption effect will be observed if $\Gamma_{inh} < \Gamma_r$.

In the case of short-lived nuclear states ($\tau < \mu$s), the requirements on times and therefore energies, are completely reversed. The interaction energy of the EM field with nuclei ($\simeq 10^{-10}$ eV) far exceeds the interaction energy of nuclei with their environment ($\simeq 10^{-14}$ eV) in a crystalline solid. Generally speaking, the process of resonant absorption can now be viewed to be instantaneous in relation to the relaxation times in the absorbing medium. This has the important consequence that in resonant absorption, the EM field encounters a snapshot of all nuclei present in a variety of local fields in the crystal. The resonance effect is crucially sensitive to inhomogeneous broadening in the lattice. This requires the more stringent condition, viz., $\Gamma_{inh} < \Gamma_n$. We are thus led to believe that the resonant absorption process from long-lived nuclear states differs qualitatively from short-lived ones. To observe these processes it is thus reasonable that the same stringent requirement, i.e., $\Gamma_{inh} < \Gamma_n$, may not be necessary in both cases as Balko *et al.* have claimed[2] in their Comment.

Mathematically, crucial differences come in. First of all, one needs to bring in fluctuations [$f(t)$ and $g(t)$] in a natural way to describe a condition of equilibrium for the case of long-lived nuclear states. In our paper,[1] we chose to define the functions $f(t)$ and $g(t)$ as generally as possible. It would be incorrect to talk about frequency and phase modulation of the wave function independently when clearly one type of modulation induces the other. In our description of the fluctuations, it would then be incorrect to assume that a random phase diffusion occur, because this would imply a decaying nuclear state, when actually what is being implied is that changes in the local chemical environment occur, inducing changes on the fields sensed by the nuclei.

Of major importance is the fact that Balko *et al.*[2] introduce correlation functions. Such correlation functions can be used to calculate ensemble averages for the case of short-lived states. Such a procedure for the case of long-lived nuclear states poses difficulties, however. It is clear that the ensemble average of the transition probabilit

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will require calculating an average over some integrals. In their Comment, Balko *et al.* proceed further by assuming that the average over the integral is equal to the integral over the average as is normally done for short-lived isotopes. This automatically leads to the definition of correlation functions. Such a procedure can only be limited to the case of short-lived nuclear states as we discuss next.

Specifically, Balko *et al.* give an expression for the probability in their Eq. (3) which reads

$$P^T(\delta\omega)=\int_0^T dt'\int_0^T dt\, f(t)f^*(t')g(t')g^*(t) \times e^{-\Gamma_a(t+t')-i\delta\omega(t-t')}. \quad (1)$$

In obtaining the ensemble average of the probability

$$\overline{\langle P^T(\delta\omega)\rangle}=\overline{\left\langle\int_0^T dt'\int_0^T dt\, f(t)f^*(t')g(t')g^*(t)e^{-\Gamma_a(t+t')-i\delta\omega(t-t')}\right\rangle} \quad (2)$$

in general, one cannot equate the average over the integrals to the integral over the average.

$$\overline{\langle P^T(\delta\omega)\rangle}=\int_0^T dt'\int_0^T dt\left\langle f(t)f^*(t')g(t')g^*(t)\right\rangle e^{-\Gamma_a(t+t')-i\delta\omega(t-t')}. \quad (3)$$

But exceptions occur, and the case of short-lived states is one of them. Indeed for short-lived states, the exponentials $e^{-\Gamma_a t'}$ and $e^{-\Gamma_a t'}$ are functions which decay much faster in time than the fluctuations $f(t)$ and $g(t)$. Therefore, contributions to the integrals in (2) over t and over t' are always limited to very short time intervals. During such short time intervals the fluctuation functions will not change appreciably and each absorption event will capture a different moment and a different possible value of these functions. In such a case one can reduce the ensemble averaged probability to an integration over the ensemble average of the fluctuating factors by introducing correlation functions, as done by Balko *et al.*[2] and others.

For long-lived isotopes, however, $e^{-\Gamma_a t'}$ and $e^{-\Gamma_a t'}$ decay so slowly that during the long emission or absorption event, $f(t)$ and $g(t)$ can fluctuate over the full range of parameter values that characterize such fluctuation functions. The procedure, followed for short-lived states, viz., sampling the fluctuating factor at the exact transition moment, can no longer be followed for long-lived states because each transition now involves a long temporal process compared to the fluctuation rates. For long-lived states, the integrations in (2) will intrinsically result in a time averaging and each nuclear event will contain the same time averaged information over these fluctuations. Because we can assume that time averaging is equal to an ensemble averaging at a given moment, there is no further need to calculate the ensemble average for the long-lived case.

Finally, if one considers the situation when a Mössbauer resonant absorption experiment is performed using a nearly monochromatic source (i.e. no fluctuations in the source matrix) to scan an absorber in which fluctuation broadening occurs, then we do expect the maximum in the resonant effect to decrease. This can be seen in Eq. (19) of Ref. 1, where if we replace the $f^{\Delta T}(\omega-\omega_0)$ function by a $\delta^{\Delta T}(\omega-\omega_0)$ function, the overlap between the source and absorber line shape decreases. The reverse situation, i.e., when the source matrix is fluctuation broadened and the absorber matrix nearly monochromatic, will likewise also give rise to a reduced maximum in resonant effect. However, if both the source and absorber matrices are fluctuation broadened in an uncorrelated manner, then our claim in Ref. 1 is that the maximum in the resonant effect will be restored.

In summary, we believe that the Comment of Balko *et al.* pertains to the usual case of the Mössbauer effect from short-lived nuclear states ($\tau \lesssim 1\mu s$). However these calculations, in our opinion, cannot be extended to the case of the Mössbauer effect from long-lived nuclear states for reasons discussed above and in our paper.[1]

[1]R. Coussement, G. S'heeren, M. Van Den Bergh, and P. Boolchand, Phys. Rev. B 45, 9755 (1992).
[2]B. Balko, I. W. Kay, J. F. Nicoll, J. D. Silk, D. A. Sparrow, and G. H. Herling, preceding paper, Phys. Rev. B 48, 16 139 (1993).

PAPER No. 4B

REPLY TO "REPLY TO 'COMMENT ON NUCLEAR RESONANT ABSORPTION IN LONG-LIVED ISOMERIC TRANSITIONS'"

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Title: Reply to "Reply to 'Comment on Nuclear resonant absorption in long-lived isomeric transitions'" by R. Coussement, G. S'heeren, M.Van en Bergh, and P. Boolchand

In their reply[1] to our comment[2] on their paper[3], the objections of the authors of References 1 and 3 were based on a conceptual argument and a mathematical concern. In simple terms, the essence of the disagreement is revealed by the answers to two questions.

(1) Conceptual question: Do long-lived and short-lived nuclear states require different treatment for describing the emission and resonance absorption processes? We say no for the problem at hand.

(2) Mathematical question: Can the average over the integral be equated to the integral over the average? We say yes, except for an integrand that is a mathematically contrived, pathological stochastic process of no interest here.

In what follows we provide a more detailed explanation of our position.

(1) Conceptual question

It has been known for some time that the nuclear lifetime affects individual level widths, but as far as fluctuations are concerned, it is an irrelevant parameter except when dealing with nonstationary stochastic processes. [4. M. Blume, *Hyperfine Structure and Nuclear Radiations*, eds. E. Matthias and D.A. Shirley, (North Holland, Amsterdam, 1968), p. 911]. Blume's theoretical treatment is completely general, as far as the relevant parameters are concerned. Both short and long lived Mössbauer isotopes are covered by the theory. Consequently, the conceptual criticism of Reference 1 is irrelevant to the issue. If there is any substance to the claim in References 1 and 3, i.e., that long-lived and short-lived states require different treatment for describing the emission and resonance absorption processes in the presence of fluctuating fields, it has to be found in the mathematical analysis.

(2) Mathematical question:

The mathematical criticism, which is the essence of the Reference 1 argument, is both confused and wrong. It is true that the time evolution of the wave function is determined by the usual quantum mechanical expression

$$\psi(t) = \exp\left[-i \int_0^t dt' \, H(t') \right] \psi(0).$$

It is the time dependent $H(t')$ that contains the stationary random process implicit in Reference 3. Although that paper characterizes the wave function as non-stationary because of the factor $e^{-\Gamma_n t/2}$, it assumes that the stochastic process represented by the factor f(t) is, in fact, stationary in connection with equation (2) of Reference 3. The representation of this time dependent operator as a factor f(t) in the wave function defined by equation (1) of the paper, i.e., in

$$\psi(x, t) = \psi(x, 0)\, f(t) e^{-\Gamma_n t/2} e^{-i\omega_0 t},$$

is an approximation whose validity must be determined in each problem. In general, f(t) need not be an example of a stationary random process since it may depend on the collective state of all of the nuclei. However, the assumptions of Reference 3 (cf. equation (2) and the surrounding discussion) are equivalent to at least a weak form of stationarity. Given stationarity of f(t), the approach used in Reference 2 is correct unless the function f(t) is pathological in nature.

Reference 1's assertion that "in general, one cannot equate the average over the integral to the integral over the average" is not applicable to the present case. On the contrary, it is well known in the theory of stochastic processes that under very general conditions:

If A is a measurable parameter set and if the average $E\{x_t(\omega)\}$ of $x_t(\omega)$ exists for $t \varepsilon A$, and

$$\int_A E\left\{\left|x_t(\omega)\right|\right\} dt < \infty,$$

then

$$E\left\{\int_A x_t(\omega) dt\right\} = \int_A E\left\{x_t(\omega)\right\} dt.$$

[5. J.L. Doob, *Stochastic Processes*, (John Wiley & Sons, New York 1953), pp. 62 ff]. In other words, for absolutely convergent iterated integrals, the result is independent of the order of integration. In less formal terms, the average of an integral can be taken over the integrand before the integration takes place, except for pathological cases. There is no evidence that suggests that the present example is such a case; however, if it were, the method proposed by R. Coussement, et al. would similarly fail.

Finally, the argument presented in the second to last paragraph of Reference 1 again ignores the distinction between overlap of lines and the resonant effect and is refuted by equation 13 in Reference 2 and the subsequent discussion.

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This document consists of a collection of papers published in the open literature in FY 1993. The first paper was published in *Phys. Rev. B* and describes the limitations of the Bonifacio-Lugiato model of superfluorescence. The second paper deals with the feasibility of nuclear superfluorescence. It was presented at the Lasers '92 Conference in Houston and published in the *Proceedings*. Paper Number 3 describes a nuclear superfluorescence model. It was presented at the Physics in Ukraine Conference in Kiev in June 1993 and published in the *Proceedings*. Paper Number 4 is a comment on a paper published in *Phys. Rev. B*, which we found to be wrong and which stated a misleading conclusion regarding the effect of homogeneous broadening on an inhomogeneously broadened system. Our comment is followed by the author's reply, which was also published in the Comments section of *Phys. Rev. B*. We include our response to the author's reply.

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